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(54) LIGHT-EMITTING DEVICE AND ILLUMINATION DEVICE USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a light-emitting device having high emission intensity by combining an exciting source radiating light having 350-415 nm wave length, with a fluorescent material.

SOLUTION: The light emitting device obtained by combining the exciting source radiating the light having 350-415 nm wave length, with the fluorescent material uses the fluorescent material containing a crystal phase having a chemical composition of general formula [1]: $M1aEubM2cM3dOe$ [1] (wherein, M1 is a metal element mixture containing Ba, Sr and Ca in a proportion of ≥ 90 mol% as the total; M2 is a metal element mixture containing Mg and Zn in a proportion of ≥ 90 mol% as the total; M3 is a metal element mixture containing Si and Ge in a proportion of ≥ 90 mol% as the total; (a) is a number satisfying the formula: $2.5 \leq (a) \leq 3.3$; b is a number satisfying the formula: $0.0001 \leq b \leq 1.0$; c is a number satisfying the formula: $0.9 \leq c \leq 1.1$; d is a number satisfying the formula: $1.8 \leq d \leq 2.2$; and e is a number satisfying the formula $7.2 \leq e \leq 8.8$).

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(54) 【発明の名称】 発光装置及びそれを用いた照明装置

(57) 【要約】

【目的】 350-415 nmの光を発生する励起源と蛍光体を組み合わせ、かつ、高い発光強度を有する発光装置を提供する。

【構成】 350-415 nmの光を発生する励起源と蛍光体を組み合わせた装置において、一般式 [1] の化学組成を有する結晶相を含有する蛍光体を使用した発光装置。

$M1aEubM2cM3d0e \cdots \cdots [1]$

但し、M1は、Ba、Sr、およびCaを合計で90mol%以上含む金属元素を表し、M2は、MgおよびZnを合計で90mol%以上含む金属元素を表し、M3は、SiおよびGeを合計で90mol%以上含む金属元素を表し、aは $2.5 \leq a \leq 3.3$ 、bは $0.0001 \leq b \leq 1.0$ 、cは $0.9 \leq c \leq 1.1$ 、dは $1.8 \leq d \leq 2.2$ 、eは $7.2 \leq e \leq 8.8$ を満足する数である。

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【特許請求の範囲】

【請求項1】 350-415nmの光を発生する第1の発光体と、当該第1の発光体からの光の照射によって可視光を発生する第2の発光体とを有する発光装置において、前記第2の発光体が、一般式〔1〕の化学組成を有する結晶相を有する蛍光体を含有してなることを特徴とする発光装置。

【化1】 $M^1aEu_bM^2cM^3dO_e \cdots \cdots$ 〔1〕

(但し、 M^1 は、Ba、Sr、およびCaからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^2 は、MgおよびZnからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^3 は、SiおよびGeからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、aは $2.5 \leq a \leq 3.3$ を満足する数、bは $0.0001 \leq b \leq 1.0$ を満足する数、cは $0.9 \leq c \leq 1.1$ を満足する数、dは $1.8 \leq d \leq 2.2$ を満足する数、eは $7.2 \leq e \leq 8.8$ を満足する数である。)

【請求項2】 Ba、Sr、およびCaの合計の M^1 中に占める割合、MgとZnの合計の M^2 中に占める割合、及びSiとGeの合計の M^3 中に占める割合が、それぞれ95mol%以上であることを特徴とする請求項1に記載の発光装置。

【請求項3】 M^1 が、Ba、Sr、およびCaから選ばれる少なくとも1種を表し、 M^2 がMgとZnから選ばれる少なくとも1種を表し、 M^3 がSiとGeから選ばれる少なくとも1種を表すことを特徴とする請求項1又は2に記載の発光装置。

【請求項4】 M^1 が、BaとSrから選ばれる少なくとも1種を表し、 M^2 がMgを表し、 M^3 がSiを表すことを特徴とする請求項1ないし3のいずれか1つに記載の発光装置。

【請求項5】 bが $0.001 \leq b \leq 1$ であることを特徴とする請求項1ないし4のいずれか1つに記載の発光装置。

【請求項6】 M^1 の中のSrに対するBaのモル比が0.05以上であることを特徴とする請求項1ないし5のいずれか1つに記載の発光装置。

【請求項7】 第1の発光体がレーザーダイオード又は発光ダイオードである請求項1乃至6のいずれか1つに記載の発光装置。

【請求項8】 第1の発光体がGaN系化合物半導体を用いたレーザーダイオード又は発光ダイオードであることを特徴とする請求項7に記載の発光装置。

【請求項9】 第1の発光体が面発光型GaN系レーザーダイオードであることを特徴とする請求項8に記載の発光装置。

【請求項10】 第2の発光体が膜状であることを特徴とする請求項9に記載の発光装置。

【請求項11】 第1の発光体の発光面に、直接第2の

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発光体の膜面を接触させてなることを特徴とする請求項1ないし10に記載の高効率な発光装置。

【請求項12】 第2の発光体が、蛍光体の粉を樹脂に分散させてなることを特徴とする請求項1ないし11のいずれか1つに記載の発光装置。

【請求項13】 請求項1ないし12の発光装置を有する照明装置。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明は発光装置に関し、詳しくは、電力源により紫外光から可視光領域の光を発光する第1の発光体と、その紫外光から可視光領域にある光を吸収し長波長の可視光を発する母体化合物が発光中心イオンを含有する蛍光体を有する波長変換材料としての第2の発光体とを組み合わせることにより、使用環境によらず演色性が良く、かつ、高強度の発光を発生させることのできる発光装置に関する。

【0002】

【従来の技術】 青、赤、緑の混色により、白色その他の様々な色を、むらなくかつ演色性良く発生させるために、LEDやLDの発光色を蛍光体で色変換させた発光装置が提案されている。例えば、特公昭49-1221号公報では、300-530nmの波長の放射ビームを発するレーザーのビームを燐光体($Y_{3-x-y}Ce_xGd_yM_{5-z}Ga_zO_{12}$ (YはY、Lu、またはLa、MはAl、Al-In、またはAl-Scを表す。))に照射させ、これを発光させてディスプレイを形成する方法が示されている。また、近年では、青色発光の半導体発光素子として注目されている発光効率の高い窒化ガリウム(GaN)系LEDやLDと、波長変換材料としての蛍光体とを組み合わせ構成される白色発光の発光装置が、消費電力が小さく長寿命であるという特徴を活かして画像表示装置や照明装置の発光源として提案されている。実際に、特開平10-242513号公報において、この窒化物系半導体のLED又はLDチップを使用し、蛍光体としてイットリウム・アルミニウム・ガーネット系を使用することを特徴とする発光装置が示されている。

【0003】 しかしながら、今までのところ、LED等の第1の発光体に対し、特開平10-242513号公報に示されるようなイットリウム・アルミニウム・ガーネット系蛍光体を第2の発光体として組み合わせた既知の発光装置では発光強度が充分とは言えず、ディスプレイやバックライト光源、信号機などの発光源としてさらなる改良が求められていた。

【0004】

【発明が解決しようとする課題】 本発明は、前述の従来技術に鑑み、発光強度の極めて高い発光装置を開発すべく、なされたものであって、従って、本発明は、製造が容易であると共に、発光強度が極めて高いダブル発光体型発光装置を得ることを提供することを目的とする。

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【0005】

【課題を解決するための手段】本発明者は、前記課題を解決すべく鋭意検討した結果、350-415nmの光を発生する第1の発光体と、当該第1の発光体からの光の照射によって可視光を発生する第2の発光体とを有する発光装置において、上記第2の発光体として下記特定の化学組成を有する結晶相を含有する蛍光体を用いると、前記蛍光体が350-415nm付近の光の照射を受け、高い強度で可視光の発光を起こす結果前記目的を達成できることを見出し本発明に到達した。即ち、本発明は、350-415nmの光を発生する第1の発光体と、当該第1の発光体からの光の照射によって可視光を発生する第2の発光体とを有する発光装置において、前記第2の発光体が、一般式[1]の化学組成を有する結晶相を有する蛍光体を含有してなることを特徴とする発光装置をその要旨とする。

【0006】

【化2】 $M^1aEu_bM^2cM^3dO_e \cdots [1]$

(但し、 M^1 は、Ba、Sr、およびCaからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^2 は、MgおよびZnからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^3 は、SiおよびGeからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、aは $2.5 \leq a \leq 3.3$ を満足する数、bは $0.0001 \leq b \leq 1.0$ を満足する数、cは $0.9 \leq c \leq 1.1$ を満足する数、dは $1.8 \leq d \leq 2.2$ を満足する数、eは $7.2 \leq e \leq 8.8$ を満足する数である。)

なお、 $Ba_3MgSi_2O_8$ 、 $Sr_3MgSi_2O_8$ 等の結晶相そのものは既知であり、これらの内のBa、SrがEu²⁺等の他の2価金属元素で置換されうること知られている。本発明は、これらのうちの $Ba_{3-x}Eu_xMgSi_2O_8$ や $Sr_{3-x}Eu_xMgSi_2O_8$ 等の化学組成を含む前記一般式[1]の化学組成を有する結晶相を有する蛍光体が、第1の発光体からの350-415nmの光の照射を受けると、他の蛍光体よりも顕著に強度の高い光を発生することを見出したことに依拠するものである。上記蛍光体は、一般的な青色発光の $BaMgAl_{10}O_{17}:Eu$ や黄色発光の $Y_3Al_5O_{12}:Ce$ よりも圧倒的に高い強度の光を発生したのである。

【0007】

【発明の実施の形態】本発明は、350-415nmの光を発生する第1の発光体と蛍光体である第2の発光体を組み合わせた発光装置であり、その第2の発光体が、下記一般式[1]の化学組成を有する結晶相を有する蛍光体を含有してなることを特徴とする。

【0008】

【化3】 $M^1aEu_bM^2cM^3dO_e \cdots [1]$

ここで、 M^1 は、Ba、Sr、およびCaからなる群か

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ら選ばれる少なくとも一種の元素を合計で90mol%以上、好ましくは95mol%以上含む金属元素を表す。中でも、 M^1 に該当する全ての元素が、Ba、Sr及びCaからなる群から選ばれた少なくとも一種であることが好ましい。その結果、より大きな発光強度を得ることができる。 M^1 として、Ba及び/又はSrを含有するのが好ましく、Baを含有するのが特に好ましい。また、Baを含有する場合、発光波長調節のし易さや発光強度等の点から、Srに対するBaのモル比は好ましくは0.05以上とするのが好ましい。この場合、Sr量は0であってもよい(この場合は上記モル比は無限大となる)が、好ましくはSrも含有させ、通常上記モル比を100以下とする。

【0009】前記一般式[1]において、 M^2 は、MgおよびZnからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上、好ましくは95mol%以上含む金属元素を表す。中でも、 M^2 に該当する全ての元素が、MgおよびZnからなる群から選ばれた少なくとも一種であることが好ましい。その結果、より大きな発光強度を得ることができる。 M^2 として、Mgを含有するのが特に好ましい。

【0010】前記一般式[1]において、 M^3 は、SiおよびGeからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上、好ましくは95mol%以上含む金属元素を表す。中でも、 M^3 に該当する全ての元素が、SiおよびGeからなる群から選ばれた少なくとも一種であることが好ましい。その結果、より大きな発光強度を得ることができる。 M^3 として、Siを含有するのが特に好ましい。

【0011】 M^1 、 M^2 、 M^3 中の金属元素として10mol%を上限、好ましくは5mol%を上限、さらに好ましくは3mol%を上限として上記以外の金属元素を結晶中に含有させることができる。この場合、その金属元素に特に制約はないが、それぞれ順にBa、Mg、Siと同じ価数、即ち2価、2価、4価の金属元素を含有させると、結晶構造を保持しやすいので好ましい。2価と4価の金属元素及び発光中心であるEu²⁺の焼成時の固体内拡散による複合酸化物の結晶化を助ける意味で、1価、3価、5価、又は6価等の金属元素を少量導入しても良い。例を挙げると、 $Ba_3MgSi_2O_8:Eu$ 蛍光体中のBa²⁺又はMg²⁺の一部を等モルのLi⁺とGa²⁺で電荷補償効果を保持しながら置換することができる。

【0012】前記一般式[1]において、a、b、c、d、及びeは、それぞれ順に M^1 に該当する金属元素のモル比、ユーロピウム原子のモル比、 M^2 に該当する金属元素のモル比、 M^3 に該当する金属元素のモル比、及び酸素原子のモル比を表す。a+b、c、d、及びeの値は、通常、概ね順に3、1、2、及び8となるが、多少のカチオン欠損や酸素欠損が多少生じていても蛍光性能に大きな影響がない等の理由により、上記の値を包含

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する許容範囲を有する。

【0013】 a は $2.5 \leq a \leq 3$ 、 3 を満足する数であるが、好ましくは 2.7 以上、より好ましくは 2.8 以上、さらに好ましくは 2.9 以上の数であり、また好ましくは 3.2 以下、さらに好ましくは 3.1 以下の数である。 b は $0.0001 \leq b \leq 1$ 、 0 を満足する数であるが、好ましくは 0.001 以上、より好ましくは 0.003 以上の数であり、また好ましくは 0.5 以下、より好ましくは 0.3 以下、さらに好ましくは 0.15 以下、特に好ましくは 0.1 以下の数である。発光中心イオンの含有量が、前記範囲未満では、発光強度が小さくなる傾向があり、一方、前記範囲超過でも、濃度消光と呼ばれる現象により、やはり発光強度が減少する傾向がある。また、結晶欠陥の少ない結晶相が得られ、発光強度が高くなる点で、 $2.7 \leq a + b \leq 3$ 、 3 を満たすことが好ましい。 c は $0.9 \leq c \leq 1$ 、 1 を満足する数であるが、好ましくは 0.93 以上、さらに好ましくは 0.95 以上の数であり、また好ましくは 1.07 以下、さらに好ましくは 1.05 以下の数である。 d は $1.8 \leq d \leq 2$ 、 2 を満足する数であるが、好ましくは 1.85 以上、さらに好ましくは 1.9 以上の数であり、また好ましくは 2.15 以下、さらに好ましくは 2.1 以下の数である。 e は $7.2 \leq e \leq 8$ 、 8 を満足する数であるが、好ましくは 7.4 以上、さらに好ましくは 7.6 以上、最も好ましくは 7.8 以上の数であり、また好ましくは 8.6 以下、さらに好ましくは 8.4 以下、最も好ましくは 8.2 以下の数である。

【0014】本発明で使用する蛍光体の典型的な結晶構造は、 $\text{Ba}_3\text{MgSi}_2\text{O}_8$ 構造、 $\text{Sr}_3\text{MgSi}_2\text{O}_8$ 構造、又は $\text{Ca}_3\text{MgSi}_2\text{O}_8$ 構造である。 $\text{Ca}_3\text{MgSi}_2\text{O}_8$ 構造は通常merwinite構造と呼ばれている。 $\text{Ba}_3\text{MgSi}_2\text{O}_8$ 構造及び $\text{Sr}_3\text{MgSi}_2\text{O}_8$ 構造は、厳密にはmerwinite構造でなく、その類似構造と考えられる。 $\text{Ba}_3\text{MgSi}_2\text{O}_8$ 構造と $\text{Sr}_3\text{MgSi}_2\text{O}_8$ 構造とは斜方晶系であり、それらの格子定数は、通常それぞれ $a=5.5\text{Å}$ 、 $b=9.8\text{Å}$ 、 $c=7.6\text{Å}$ 、及び $a=5.4\text{Å}$ 、 $b=9.6\text{Å}$ 、 $c=7.2\text{Å}$ 程度である。 $\text{Ca}_3\text{MgSi}_2\text{O}_8$ 構造は、単斜晶系、空間群 $P2_1/a$ であり、格子定数は通常、 $a=13.254\text{Å}$ 、 $b=5.293\text{Å}$ 、 $c=9.328\text{Å}$ 程度である。図1、2、及び3に、それぞれ $\text{Ba}_3\text{MgSi}_2\text{O}_8$ 、 $\text{Sr}_3\text{MgSi}_2\text{O}_8$ 、及び $\text{Ca}_3\text{MgSi}_2\text{O}_8$ のX線回折パターンを示す(粉末X線回折データベースより)。これらの結晶構造においては、Ba、Sr、Ca、その他の2価金属は互いが固溶し合う組成範囲が広いので、構造が近いと考えられる。本発明で使用する蛍光体の結晶相は、通常 $\text{Ba}_3\text{MgSi}_2\text{O}_8$ 構造、 $\text{Sr}_3\text{MgSi}_2\text{O}_8$ 構造、又は $\text{Ca}_3\text{MgSi}_2\text{O}_8$ 構造を持つ物質に対し付活剤として Eu^{2+} を置換させたものに該当する。

【0015】本発明で使用する蛍光体は、第1の発光体からの $350\sim415\text{nm}$ の光によって励起され、可視光を発生する。上記蛍光体は、 $350\sim415\text{nm}$ の光の励起によって非常に強い発光強度の可視光を発生す

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る。本発明で使用する蛍光体は、前記一般式[1]に示されるような M^1 源、 M^2 源、 M^3 源の化合物、並びに、発光中心イオンの元素(Eu)源化合物を、ハンマーミル、ローミル、ボールミル、ジェットミル等の乾式粉碎機を用いて粉碎した後、リボンブレンダー、V型ブレンダー、ヘンシェルミキサー等の混合機により混合するか、或いは、混合した後、乾式粉碎機を用いて粉碎する乾式法、又は、水等の媒体中にこれらの化合物を加え、媒体攪拌式粉碎機等の湿式粉碎機を用いて粉碎及び混合するか、或いは、これらの化合物を乾式粉碎機により粉碎した後、水等の媒体中に加え混合することにより調製されたスラリーを、噴霧乾燥等により乾燥させる湿式法により、調製した粉碎混合物を、加熱処理して焼成することにより製造することができる。

【0016】これらの粉碎混合法の中で、特に、発光中心イオンの元素源化合物においては、少量の化合物を全体に均一に混合、分散させる必要があることから液体媒体を用いるのが好ましく、又、他の元素源化合物において全体に均一な混合が得られる面からも、後者湿式法が好ましく、又、加熱処理法としては、アルミナや石英製の坩堝やトレイ等の耐熱容器中で、通常 $800\sim1600^\circ\text{C}$ 、好ましくは $1000\sim1400^\circ\text{C}$ の温度で、大気、酸素、一酸化炭素、二酸化炭素、窒素、水素、アルゴン等の気体の単独或いは混合雰囲気下、 $10\text{分}\sim24$ 時間、加熱することによりなされる。尚、加熱処理後、必要に応じて、洗浄、乾燥、分級処理等がなされる。

【0017】尚、前記加熱雰囲気としては、発光中心イオンの元素が発光に寄与するイオン状態(価数)を得るために必要な雰囲気が選択される。本発明における2価の Eu 等の場合には、一酸化炭素、窒素、水素、アルゴン等の中性若しくは還元雰囲気下が好ましいが、大気、酸素等の酸化雰囲気下も条件さえ選べば可能である。又、ここで、 M^1 源、 M^2 源、及び M^3 源の化合物、並びに、発光中心イオンの元素源化合物としては、 M^1 、 M^2 、及び M^3 、並びに発光中心イオンの元素の各酸化物、水酸化物、炭酸塩、硝酸塩、硫酸塩、蔞酸塩、カルボン酸塩、ハロゲン化物等が挙げられ、これらの中から、複合酸化物への反応性、及び、焼成時における NO_x 、 SO_x 等の非発生性等を考慮して選択される。

【0018】金属元素 M^1 に対して好ましいとする前記Ba、Sr、及びCaについて、それらの M^1 源化合物を具体的に例示すれば、Ba源化合物としては、 BaO 、 $\text{Ba(OH)}_2 \cdot 8\text{H}_2\text{O}$ 、 BaCO_3 、 $\text{Ba(NO}_3)_2$ 、 BaSO_4 、 $\text{Ba(OCO)}_2 \cdot 2\text{H}_2\text{O}$ 、 $\text{Ba(OCOCH}_3)_2$ 、 BaCl_2 等が、又、Sr源化合物としては、 SrO 、 $\text{Sr(OH)}_2 \cdot 8\text{H}_2\text{O}$ 、 SrCO_3 、 $\text{Sr(NO}_3)_2$ 、 SrSO_4 、 $\text{Sr(OCO)}_2 \cdot \text{H}_2\text{O}$ 、 $\text{Sr(OCOCH}_3)_2 \cdot 0.5\text{H}_2\text{O}$ 、 SrCl_2 等が、又、Ca源化合物としては、 CaO 、 Ca(OH)_2 、 CaCO_3 、 $\text{Ca(NO}_3)_2 \cdot 4\text{H}_2\text{O}$ 、 CaSO_4 ・

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$2\text{H}_2\text{O}$ 、 $\text{Ca}(\text{OCO})_2 \cdot \text{H}_2\text{O}$ 、 $\text{Ca}(\text{OCOCH}_3)_2 \cdot \text{H}_2\text{O}$ 、 CaCl_2 等がそれぞれ挙げられる。

【0019】又、金属元素 M^2 に対して好ましいとする前記 Mg 及び Zn について、それらの M^2 源化合物を具体的に例示すれば、 Mg 源化合物としては、 MgO 、 $\text{Mg}(\text{OH})_2$ 、 MgCO_3 、 $\text{Mg}(\text{OH})_2 \cdot 3\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$ 、 $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ 、 MgSO_4 、 $\text{Mg}(\text{OCO})_2 \cdot 2\text{H}_2\text{O}$ 、 $\text{Mg}(\text{OCOCH}_3)_2 \cdot 4\text{H}_2\text{O}$ 、 MgCl_2 等が、又、 Zn 源化合物としては、 ZnO 、 $\text{Zn}(\text{OH})_2$ 、 ZnCO_3 、 $\text{Zn}(\text{NO}_3)_2$ 、 $\text{Zn}(\text{OCO})_2$ 、 $\text{Zn}(\text{OCOCH}_3)_2$ 、 ZnCl_2 等がそれぞれ挙げられる。

【0020】又、金属元素 M^3 に対して好ましいとする前記 Si 及び Ge について、それらの M^3 源化合物を具体的に例示すれば、 Si 源化合物としては、 SiO_2 、 H_4SiO_4 、 $\text{Si}(\text{OCOCH}_3)_4$ 等が、又、 Ge 源化合物としては、 GeO_2 、 $\text{Ge}(\text{OH})_4$ 、 $\text{Ge}(\text{OCOCH}_3)_4$ 、 GeCl_4 等がそれぞれ挙げられる。更に、発光中心イオンの元素として好ましいとする前記 Eu について、その元素源化合物を具体的に例示すれば、 Eu_2O_3 、 $\text{Eu}_2(\text{SO}_4)_3$ 、 $\text{Eu}_2(\text{OCO})_6$ 、 EuCl_2 、 EuCl_3 等が挙げられる。

【0021】本発明において、前記蛍光体に光を照射する第1の発光体は、波長 $350-415\text{nm}$ の光を発生する。好ましくは波長 $350-415\text{nm}$ の範囲にピーク波長を有する光を発生する発光体を使用する。第1の発光体の具体例としては、発光ダイオード(LED)またはレーザーダイオード(LD)等を挙げることができる。特に消費電力を抑制できる点でレーザーダイオードが好ましい。また、 GaN 系化合物半導体を使用した、 GaN 系LEDやLDが好ましい。なぜなら、 GaN 系LEDやLDは、この領域の光を発する SiC 系LED等と比し、発光出力や外部量子効率が格段に大きく、前記蛍光体と組み合わせることによって、非常に低電力で非常に明るい発光が得られるからである。例えば、 20mA の電流負荷に対し、通常 GaN 系は SiC 系の10倍以上の発光強度を有する。 GaN 系LEDやLDにおいては、 $\text{Al}_x\text{Ga}_{1-x}\text{N}$ 発光層、 GaN 発光層、または $\text{In}_x\text{Ga}_{1-x}\text{N}$ 発光層を有しているものが好ましい。 GaN 系LEDにおいては、それらの中で $\text{In}_x\text{Ga}_{1-x}\text{N}$ 発光層を有するものが発光強度が非常に強いので、特に好ましく、 GaN 系LDにおいては、 $\text{In}_x\text{Ga}_{1-x}\text{N}$ 層と GaN 層の多重量子井戸構造のものが発光強度が非常に強いので、特に好ましい。なお、上記において $x+y$ の値は通常 $0.8 \sim 1.2$ の範囲の値である。 GaN 系LEDにおいて、これら発光層に Zn や Si をドーパしたものとドーパント無しとのものが発光特性を調節する上で好ましいものである。 GaN 系LEDはこれら発光層、 p 層、 n 層、電極、および基板を基本構成要素としたものであり、発光層を n 型と p 型の $\text{Al}_x\text{Ga}_{1-x}\text{N}$ 層、 GaN 層、または $\text{In}_x\text{Ga}_{1-x}\text{N}$ 層などで

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サンドイッチにしたヘテロ構造を有しているものが発光効率が高く、好ましく、さらにヘテロ構造を量子井戸構造にしたものが発光効率がさらに高く、より好ましい。

【0022】本発明においては、面発光型の発光体、特に面発光型 GaN 系レーザーダイオードを第1の発光体として使用することは、発光装置全体の発光効率を高めることになるので、特に好ましい。面発光型の発光体とは、膜の面方向に強い発光を有する発光体であり、面発光型 GaN 系レーザーダイオードにおいては、発光層等の結晶成長を制御し、かつ、反射層等をうまく工夫することにより、発光層の縁方向よりも面方向の発光を強くすることができる。面発光型のものを使用することによって、発光層の縁から発光するタイプに比べ、単位発光量あたりの発光断面積が大きくとれる結果、第2の発光体の蛍光体にその光を照射する場合、同じ光量で照射面積を非常に大きくすることができ、照射効率を良くすることができるので、第2の発光体である蛍光体からより強い発光を得ることができる。

【0023】第1の発光体として面発光型のものを使用する場合、第2の発光体を膜状とするのが好ましい。その結果、面発光型の発光体からの光は断面積が十分大きいので、第2の発光体をその断面の方向に膜状とすると、第1の発光体からの蛍光体への照射断面積が蛍光体単位量あたり大きくなるので、蛍光体からの発光の強度をより大きくすることができる。

【0024】また、第1の発光体として面発光型のものを使用し、第2の発光体として膜状のものをを用いる場合、第1の発光体の発光面に、直接膜状の第2の発光体を接触させる形状とするのが好ましい。ここでいう接触とは、第1の発光体と第2の発光体とが空気や気体を介さないでぴたりと接している状態をつくることを言う。その結果、第1の発光体からの光が第2の発光体の膜面で反射されて外にしみ出るといった光量損失を避けることができるので、装置全体の発光効率を良くすることができる。

【0025】本発明の発光装置の一例における第1の発光体と第2の発光体との位置関係を示す模式的斜視図を図4に示す。図4中の1は、前記蛍光体を有する膜状の第2の発光体、2は第1の発光体としての面発光型 GaN 系LD、3は基板を表す。相互に接触した状態をつくるために、LD2と第2の発光体1とそれぞれ別個にをつくっておいてそれらの面同士を接着剤やその他の手段によって接触させても良いし、LD2の発光面上に第2の発光体をを製膜(成型)させても良い。これらの結果、LD2と第2の発光体1とを接触した状態とすることができる。

【0026】第1の発光体からの光や第2の発光体からの光は通常四方八方に向いているが、第2の発光体の蛍光体の粉を樹脂中に分散させると、光が樹脂の外に出る時にその一部が反射されるので、ある程度光の向きを揃

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えられる。従って、効率の良い向きに光をある程度誘導できるので、第2の発光体として、前記蛍光体の粉を樹脂中へ分散したものを使用するのが好ましい。また、蛍光体を樹脂中に分散させると、第1の発光体からの光の第2の発光体への全照射面積が大きくなるので、第2の発光体からの発光強度を大きくすることができるという利点も有する。この場合に使用できる樹脂としては、エポキシ樹脂、ポリビニル系樹脂、ポリエチレン系樹脂、ポリプロピレン系樹脂、ポリエステル系樹脂等各種のものが挙げられるが、蛍光体粉の分散性が良い点で好ましくはエポキシ樹脂である。第2の発光体の粉を樹脂中に分散させる場合、当該第2の発光体の粉と樹脂の全体に対するその粉の重量比は、通常10~95%、好ましくは20~90%、さらに好ましくは30~80%である。蛍光体が多すぎると粉の凝集により発光効率が低下することがあり、少なすぎると今度は樹脂による光の吸収や散乱のため発光効率が低下することがある。

【0027】本発明の発光装置は、波長変換材料としての前記蛍光体と、350~415 nmの光を発生する発光素子とから構成されてなり、前期蛍光体が発光素子の発する350~415 nmの光を吸収して、使用環境によらず演色性が良く、かつ、高強度の可視光を発生させることのできる発光装置であり、バックライト光源、信号機などの発光源、又、カラー液晶ディスプレイ等の画像表示装置や面発光等の照明装置等の光源に適している。

【0028】本発明の発光装置を図面に基づいて説明すると、図5は、第1の発光体(350~415 nm発光体)と第2の発光体とを有する発光装置の一実施例を示す模式的断面図であり、4は発光装置、5はマウントリード、6はインナーリード、7は第1の発光体(350~415 nmの発光体)、8は第2の発光体としての蛍光体含有樹脂部、9は導電性ワイヤー、10はモールド部材である。

【0029】本発明の一例である発光装置は、図5に示されるように、一般的な砲弾型の形態をなし、マウントリード5の上部カップ内には、Ga N系発光ダイオード等からなる第1の発光体(350~415 nm発光体)7が、その上に、蛍光体をエポキシ樹脂やアクリル樹脂等のバインダーに混合、分散させ、カップ内に流し込むことにより第2の発光体として形成された蛍光体含有樹脂部8で被覆されることにより固定されている。一方、第1の発光体7とマウントリード5、及び第1の発光体7とインナーリード6は、それぞれ導電性ワイヤー9で導通されており、これら全体がエポキシ樹脂等によるモールド部材10で被覆、保護されてなる。

【0030】又、この発光素子1を組み込んだ面発光照明装置98は、図9に示されるように、内面を白色の平滑面等の光不透過性とした方形の保持ケース910の底面に、多数の発光装置91を、その外側に発光素子91

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の駆動のための電源及び回路等(図示せず。)を設けて配置し、保持ケース910の蓋部に相当する箇所、乳白色としたアクリル板等の拡散板99を発光の均一化のために固定してなる。

【0031】そして、面発光照明装置98を駆動して、発光素子91の第1の発光体に電圧を印加することにより350~415 nmの光を発光させ、その発光の一部を、第2の発光体としての蛍光体含有樹脂部における前記蛍光体が吸収し、可視光を発光し、一方、蛍光体に吸収されなかった青色光等との混色により演色性の高い発光が得られ、この光が拡散板99を透過して、図面上方に射出され、保持ケース910の拡散板99面内において均一な明るさの照明光が得られることとなる。

【0032】

【実施例】以下、本発明を実施例によりさらに具体的に説明するが、本発明はその要旨を越えない限り以下の実施例に限定されるものではない。なお、相対強度は、比較例1の蛍光体の発光強度を100としたときの発光強度の大きさを示す。

実施例1

M¹源化合物としてBaCO₃; 0.0553モル、M²源化合物として塩基性炭酸マグネシウム(Mgのモル数0.0186モル)、及びM³源化合物としてSiO₂; 0.0372モル、並びに発光中心イオンの元素源化合物としてEu₂O₃; 0.00018モルを純水と共に、アルミナ製容器及びビーズの湿式ボールミル中で粉碎、混合し、乾燥後、ナイロンメッシュを通過させた後、得られた粉碎混合物をアルミナ製坩堝中で、4%の水素を含む窒素ガス流下、1200℃で2時間、加熱することにより焼成し、引き続いて、水洗浄、乾燥、及び分級処理を行うことにより蛍光体を製造した。

【0033】図6に、得られた蛍光体Ba_{2.98}Eu_{0.02}MgSi₂O₈のX線回折パターンを示す。図6のピークパターンは図1のBa₃MgSi₂O₈のそれと結晶構造的に一致していることがわかる。図7に、Ga N系発光ダイオードの紫外光領域の主波長である400 nmでこの蛍光体を励起したときの発光スペクトルを示す。表1にその発光ピークの波長と相対強度を示す。

【0034】実施例2

M¹源化合物としてSrCO₃; 0.0553モルを用いた外は、実施例1と同様にして蛍光体を製造した。図8に、この蛍光体Sr_{2.98}Eu_{0.02}MgSi₂O₈のX線回折パターンを示す。図8のピークパターンは図2のSr₃MgSi₂O₈のそれと結晶構造的に一致していることがわかる。表1に、Ga N系発光ダイオードの紫外光領域の主波長である400 nmでこの蛍光体を励起したときの発光ピークの波長と相対強度を示す。

【0035】実施例3

M¹源化合物としてBaCO₃; 0.0442モル、CaCO₃; 0.0084モル、及びMnCO₃; 0.002

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8モルを用いた外は、実施例1と同様にして蛍光体を製造した。表-1に、Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長と相対強度を示す。

【0036】実施例4

M²源化合物として塩基性炭酸亜鉛（Znのモル数0.0186モル）を用いた外は、実施例1と同様にして蛍光体を製造した。表-1に、Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長と相対強度を示す。

【0037】実施例5

発光中心イオンの元素源化合物であるEu₂O₃のモル数を0.000074モルと変え、M¹源化合物としてBaCO₃のモル数を0.0556モルと変えた以外は、実施例1と同様にして蛍光体を製造した。表-1に、Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長と相対強度を示す。

【0038】実施例6

焼成温度を1300℃と変えた以外は、実施例1と同様にして蛍光体を製造した。表-1に、Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長と相対強度を示した。

【0039】実施例7

BaCO₃の配合量を0.0549モルに、Eu₂O₃の配合量を0.00047モルに変えた以外は、実施例1と同様にして蛍光体Ba₂.₉₅Eu_{0.05}MgSi₂O₈を製造した。Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長は438nm、相対強度は299であった。

【0040】実施例8

BaCO₃の配合量を0.0539モルに、Eu₂O₃の配合量を0.00093モルに変えた以外は、実施例1と同様にして蛍光体Ba₂.₉Eu_{0.1}MgSi₂O₈を製造した。Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長は440nm、相対強度は320であった。

【0041】実施例9

BaCO₃の配合量を0.0530モルに、Eu₂O₃の配合量を0.00140モルに変えた以外は、実施例1と同様にして蛍光体Ba₂.₈₅Eu_{0.15}MgSi₂O₈を製造した。Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長は440、相対強度は261であった。

【0042】実施例10

BaCO₃の配合量を0.0521モルに、Eu₂O₃

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の配合量を0.00186モルに変えた以外は、実施例1と同様にして蛍光体Ba₂.₈Eu_{0.2}MgSi₂O₈を製造した。Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長は440、相対強度は199であった。

【0043】実施例11

BaCO₃の配合量を0.0502モルに、Eu₂O₃の配合量を0.00279モルに変えた以外は、実施例1と同様にして蛍光体Ba₂.₇Eu_{0.3}MgSi₂O₈を製造した。Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光ピークの波長は441、相対強度は113であった。

【0044】比較例1

BaCO₃; 0.0103モル、塩基性炭酸マグネシウム（Mgのモル数0.0103モル）、及びγ-Al₂O₃; 0.0570モル、並びに発光中心イオンの元素源化合物としてEu₂O₃; 0.00057モルを純水と共に、アルミナ製容器及びビーズの湿式ボールミル中で粉碎、混合し、乾燥後、ナイロンメッシュを通過させた後、得られた粉碎混合物をアルミナ製坩堝中で、4%の水素を含む窒素ガス流下、1500℃で2時間、加熱することにより焼成し、引き続いて、水洗浄、乾燥、及び分級処理を行うことにより青色発光の蛍光体（Ba_{0.9}Eu_{0.1}MgAl₁₀O₁₇）を製造した。図7に、Ga₂N系発光ダイオードの紫外光領域の主波長である400nmでこの蛍光体を励起したときの発光スペクトルを示し、実施例1と比較例1の青色発光蛍光体の性能を比較した。表-1にその発光ピークの波長と相対強度を示した。400nm励起による実施例1の蛍光体の発光強度が比較例1の蛍光体のその2.8倍もあることがわかる。

【0045】比較例2

Y₂O₃; 0.0238モル、γ-Al₂O₃; 0.0400モル、並びに発光中心イオンの元素源化合物としてCeO₂; 0.00048モルを純水と共に、アルミナ製容器及びビーズの湿式ボールミル中で粉碎、混合し、乾燥後、ナイロンメッシュを通過させた後、得られた粉碎混合物をアルミナ製坩堝中で、4%の水素を含む窒素ガス流下、1500℃で2時間、加熱することにより焼成し、引き続いて、水洗浄、乾燥、及び分級処理を行うことにより黄色発光の蛍光体（Y₂.₉₈Ce_{0.03}Al₅O₁₂）を製造した。表-1にその発光ピークの波長と相対強度を示した。400nm励起による実施例1の蛍光体の発光強度が比較例2の蛍光体のその250倍もあることがわかる。

【0046】

【表1】

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表-1

実施例又は比較例	蛍光体の化学組成	発光ピーク波長 (nm)	相対発光強度
実施例1	Ba ₂ ...Eu...MgSi ₂ O ₈	434	277
実施例2	Sr...Eu...MgSi ₂ O ₈	455	238
実施例3	Ba...Eu...Ca...Mn...MgSi ₂ O ₈	440	227
実施例4	Ba...Eu...ZnSi ₂ O ₈	436	255
実施例5	Ba...Eu...MgSi ₂ O ₈	435	225
実施例6	Ba...Eu...MgSi ₂ O ₈	434	277
比較例1	Ba...Eu...MgAl ₂ O ₄	448	100
比較例2	Y...Ce...Al ₂ O ₃	561	1.1

【0047】

【発明の効果】本発明によれば、発光強度の高い発光装置を提供することができる。

【図面の簡単な説明】

【図1】 Ba₃MgSi₂O₈のX線回折パターン(X線源Cu Kαに換算したもの)

【図2】 Sr₃MgSi₂O₈のX線回折パターン(X線源Cu Kαに換算したもの)

【図3】 Ca₃MgSi₂O₈のX線回折パターン(X線源Cu Kαに換算したもの)

【図4】 本発明の発光装置の一例における第1の発光体と第2の発光体との位置関係を示す模式的斜視図。

【図5】、第1の発光体(350-415nm発光体)と第2の発光体とを有する発光装置の一実施例を示す模式的断面図。

【図6】 実施例1の蛍光体のX線回折パターン(X線源：Cu Kα)

【図7】 発光波長400nmのGaN系発光ダイオード

に本発明の実施例1と比較例1の蛍光体をそれぞれ組み合わせたときの発光スペクトル。

【図8】 実施例2の蛍光体のX線回折パターン(X線源：Cu Kα)

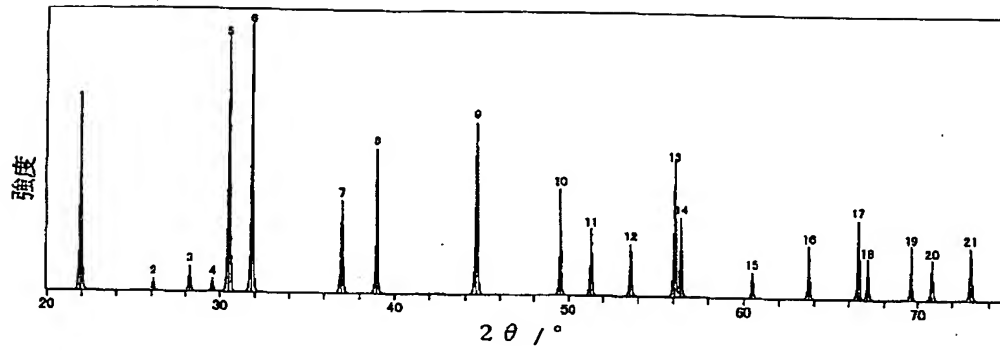
【図9】 本発明の面発光照明装置の一例を示す模式的断面図。

【符号の説明】

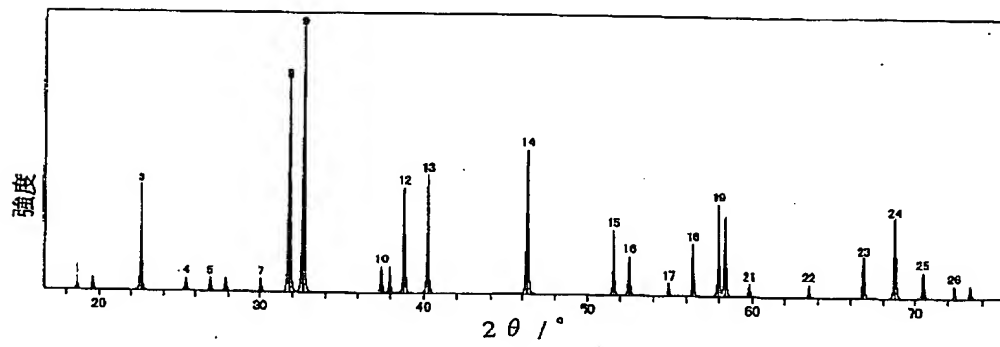
- 1 ; 第2の発光体
- 2 ; 面発光型GaN系LED
- 3 ; 基板
- 4 ; 発光装置
- 5 ; マウントリード
- 6 ; インナーリード
- 7 ; 第1の発光体(350~415nmの発光体)
- 8 ; 本発明中の蛍光体を含有させた樹脂部
- 9 ; 導電性ワイヤー
- 10 ; モールド部材

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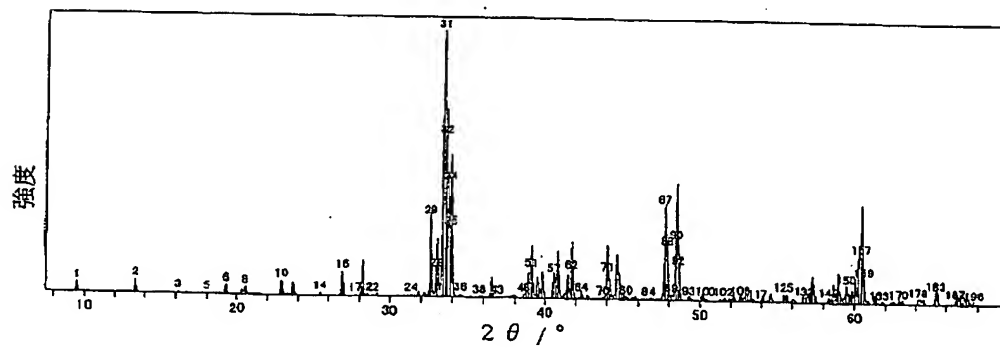
【図1】



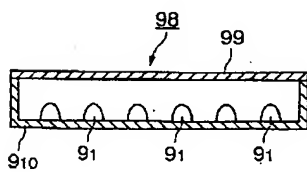
【図2】



【図3】

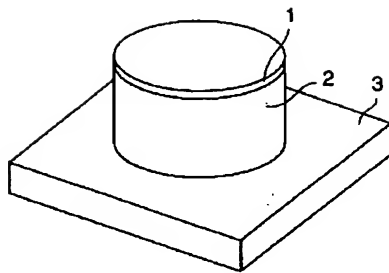


【図9】

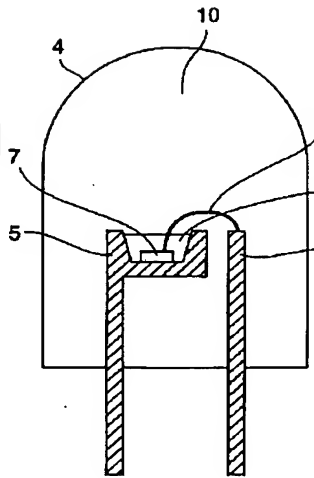


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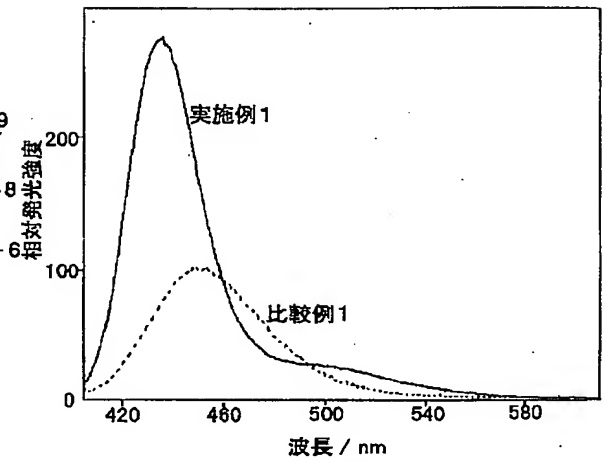
【図4】



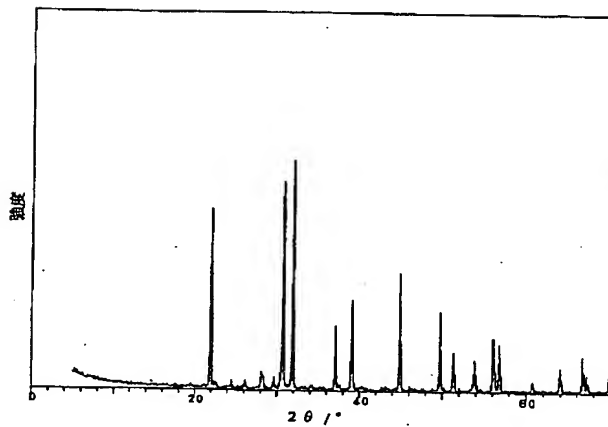
【図5】



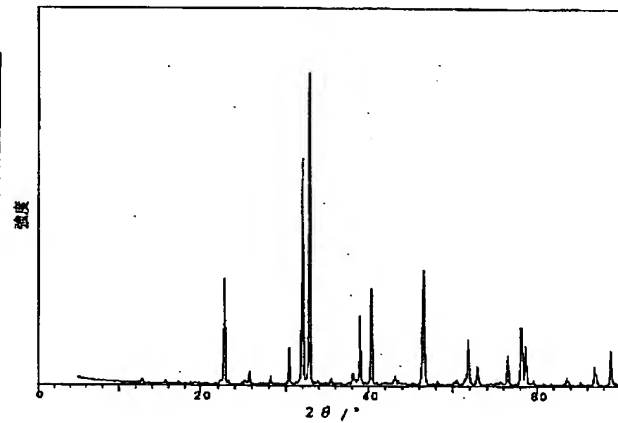
【図7】



【図6】



【図8】



フロントページの続き

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【手続補正書】

【提出日】2005年(2005)12月9日

【手続補正1】

【補正対象書類名】明細書

【補正対象項目名】特許請求の範囲

【補正方法】変更

【補正の内容】

【特許請求の範囲】

【請求項1】 350-415nmの光を発生する第1の発光体と、当該第1の発光体からの光の照射によって可視光を発生する第2の発光体とを有する発光装置において、前記第2の発光体が、一般式[1]の化学組成を有する結晶相を有する蛍光体を含有してなることを特徴とする発光装置。

【化1】



(但し、 M^1 は、Ba、Sr、およびCaからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^2 は、MgおよびZnからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 M^3 は、SiおよびGeからなる群から選ばれる少なくとも一種の元素を合計で90mol%以上含む金属元素を表し、 a は $2.5 \leq a \leq 3.3$ を満足する数、 b は $0.0001 \leq b \leq 1.0$ を満足する数、 c は $0.9 \leq c \leq 1.1$ を満足する数、 d は $1.8 \leq d \leq 2.2$ を満足する数、 e は $7.2 \leq e \leq 8.8$ を満足する数である。)

【請求項2】 Ba、Sr、およびCaの合計の M^1 中に占める割合、MgとZnの合計の M^2 中に占める割合、及びSiとGeの合計の M^3 中に占める割合が、それぞれ95mol%以上であることを特徴とする請求項1に記載の発光装置。

【請求項3】 M^1 が、Ba、Sr、およびCaから選ばれる少なくとも1種を表し、 M^2 がMgとZnから選ばれる少なくとも1種を表し、 M^3 がSiとGeから選ばれる少なくとも1種を表すことを特徴とする請求項1又は2に記載の発光装置。

【請求項4】 M^1 が、BaとSrから選ばれる少なくとも1種を表し、 M^2 がMgを表し、 M^3 がSiを表すことを特徴とする請求項1ないし3のいずれか1項に記載の発光装置。

【請求項5】 b が $0.001 \leq b \leq 1$ であることを特徴とする請求項1ないし4のいずれか1項に記載の発光装置。

【請求項6】 M^1 の中のSrに対するBaのモル比が0.05以上であることを特徴

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とする請求項1ないし5のいずれか1項に記載の発光装置。

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【請求項7】 第1の発光体がレーザーダイオード又は発光ダイオードであることを特徴とする請求項1乃至6のいずれか1項に記載の発光装置。

【請求項8】 第1の発光体がGaN系化合物半導体を用いたレーザーダイオード又は発光ダイオードであることを特徴とする請求項7に記載の発光装置。

【請求項9】 第1の発光体が面発光型GaN系レーザーダイオードであることを特徴とする請求項8に記載の発光装置。

【請求項10】 第2の発光体が膜状であることを特徴とする請求項9に記載の発光装置。

【請求項11】 第1の発光体の発光面に、直接第2の発光体の膜面を接触させてなることを特徴とする請求項1ないし10のいずれか1項に記載の高効率な発光装置。

【請求項12】 第2の発光体が、蛍光体の粉を樹脂に分散させてなることを特徴とする請求項1ないし11のいずれか1項に記載の発光装置。

【請求項13】 請求項1ないし12のいずれか1項に記載の発光装置を有する照明装置。

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CLAIMS

[Claim(s)]

[Claim 1] Luminescence equipment characterized by coming to contain the fluorescent substance which has the crystal phase in which said 2nd emitter has the chemical composition of a general formula [1] in the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned.

[Formula 1] $M1aEubM2cM3dOe$ [1]

The included metallic element is expressed, however, at least a kind of element chosen from the group which M1 becomes from Ba, Sr, and calcium — the sum total — more than 90mol% — M2 The included metallic element is expressed, at least a kind of element chosen from the group which consists of Mg and Zn — the sum total — more than 90mol% — M3 The included metallic element is expressed, at least a kind of element chosen from the group which consists of Si and germanium — the sum total — more than 90mol% — the number with which a is satisfied of $2.5 \leq a \leq 3.3$, the number with which b is satisfied of $0.0001 \leq b \leq 1.0$, the number with which c is satisfied of $0.9 \leq c \leq 1.1$, the number with which d is satisfied of $1.8 \leq d \leq 2.2$, and e are numbers with which are satisfied of $7.2 \leq e \leq 8.8$.

[Claim 2] Luminescence equipment according to claim 1 with which the rate of occupying in M1 of the sum total of Ba, Sr, and calcium, the rate of occupying in M2 of the sum total of Mg and Zn, and the rate of occupying in M3 of the sum total of Si and germanium are characterized by being more than 95mol%, respectively.

[Claim 3] Luminescence equipment according to claim 1 or 2 characterized by for M1 expressing at least one sort chosen from Ba, Sr, and calcium, expressing at least one sort as which M2 is chosen from Mg and Zn, and expressing at least one sort as which M3 is chosen from Si and germanium.

[Claim 4] Claim 1 characterized by for M1 expressing at least one sort chosen from Ba and Sr, for M2 expressing Mg, and M3 expressing Si thru/or luminescence equipment of any one publication of three.

[Claim 5] Claim 1 characterized by b being $0.001 \leq b \leq 1$ thru/or luminescence equipment of any one publication of four.

[Claim 6] Claim 1 characterized by the mole ratio of Ba to Sr in M1 being 0.05 or more thru/or luminescence equipment of any one publication of five.

[Claim 7] Claim 1 whose 1st emitter is laser diode or light emitting diode thru/or luminescence equipment of any one publication of six.

[Claim 8] Luminescence equipment according to claim 7 with which the 1st emitter is characterized by being the laser diode or light emitting diode using a GaN system compound semiconductor.

[Claim 9] Luminescence equipment according to claim 8 characterized by the 1st emitter being surface-emitting type GaN system laser diode.

[Claim 10] Luminescence equipment according to claim 9 characterized by the 2nd emitter being the film-like.

[Claim 11] Claim 1 characterized by making the film surface of the 2nd emitter come to contact the luminescence side of the 1st emitter directly thru/or efficient luminescence equipment given in 10.

[Claim 12] Claim 1 to which the 2nd emitter is characterized by making resin come to distribute the powder of a fluorescent substance thru/or luminescence equipment of any one publication of 11.

[Claim 13] The lighting system which has claim 1 thru/or luminescence equipment of 12.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] The 1st emitter to which this invention emits light in the light of a light field from ultraviolet radiation by the power source in detail about luminescence equipment. By combining the 2nd emitter which has the fluorescent substance with which the parent compound which absorbs the light which is in a light field from the ultraviolet radiation, and emits the light of long wavelength contains emission center ion and which carries out a wavelength conversion ingredient It is not based on an operating environment, but color rendering properties are good and are related with the luminescence equipment which can be made to generate luminescence of high intensity.

[0002]

[Description of the Prior Art] The luminescence equipment which carried out color conversion of the luminescent color of LED or LD with the fluorescent substance with blue, red, and green color mixture in order to generate various colors of white and others with color rendering properties uniform and sufficient is proposed. For example, by JP,49-1221,B, the beam of the laser which emits the radiation beam of the wavelength of 300 to 530 nm is made to irradiate a phosphor (Y₃-x-yCexGdyM₅-zGazO₁₂ (for Y to express La and for Y, Lu, or M to express aluminum, aluminum-In, or aluminum-Sc.)), and the approach of making this emit light and forming a display is shown. Moreover, the luminescence equipment of white luminescence which consists of recent years combining the gallium nitride (GaN) systems LED and LD with high luminous efficiency and the fluorescent substance as a wavelength conversion ingredient which attract attention as a semi-conductor light emitting device of blue luminescence is proposed as a source of luminescence of an image display device or a lighting system taking advantage of the description that power consumption is small and it is long lasting. The luminescence equipment characterized by using LED or LD chip of this nitride system semi-conductor, and actually using an yttrium aluminum garnet system as a fluorescent substance in JP,10-242513,A is shown.

[0003] However, despite former, it could not say that the known luminescence equipment which combined the yttrium aluminum garnet system fluorescent substance as shown in JP,10-242513,A as the 2nd emitter was enough as luminescence reinforcement to the 1st emitter, such as LED, but the further amelioration was called for as sources of luminescence, such as a display, and the back light light source, a signal.

[0004]

[Problem(s) to be Solved by the Invention] This invention is made in view of the above-mentioned conventional technique that the very high luminescence equipment of luminescence reinforcement should be developed, therefore this invention aims at offering obtaining double emitter mold luminescence equipment with very high luminescence reinforcement while it is easy to manufacture.

[0005]

[Means for Solving the Problem] In the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned as a result of inquiring wholeheartedly that this invention person should solve said technical problem When the fluorescent substance containing the crystal phase which has the chemical composition of the following specification as the 2nd emitter of the above was used, said fluorescent substance received the exposure of the light near 350-415nm, it found out that said object could be attained as a result of causing luminescence of the light by high reinforcement, and this invention was reached. That is, let the luminescence equipment characterized by this invention coming to contain the fluorescent substance which has the crystal phase in which said 2nd emitter has the chemical composition of a general formula [1] in the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned be the summary.

[0006]

[Formula 2] M₁aEubM₂cM₃dO_e [1]

The included metallic element is expressed. however, at least a kind of element chosen from the group which M₁ becomes from Ba, Sr, and calcium -- the sum total -- more than 90mol% -- M₂ The included metallic element is expressed. at least a kind of element chosen from the group which consists of Mg and Zn -- the sum total -- more than 90mol% -- M₃ The included metallic element is expressed. at least a kind of element chosen from the group which consists of Si and germanium -- the sum total -- more than 90mol% -- the number with which a is satisfied of 2.5<=a<=3.3, the number with which b is satisfied of 0.0001<=b<=1.0, the number with which c is satisfied of 0.9<=c<=1.1, the number with which d is satisfied of 1.8<=d<=2.2, and e are numbers with which are satisfied of 7.2<=e<=8.8.

In addition, the crystal phase of Ba₃MgSi₂O₈ and Sr₃MgSi₂O₈ grade itself is known, and it is also known that Ba of these and Sr will be permuted by other divalent metal elements, such as Eu²⁺, and will get. This invention is based on having found out generating light with notably high reinforcement rather than other fluorescent substances, if the fluorescent substance which has the crystal phase which has the chemical composition of said general formula [1] including the chemical composition of Ba₃-xEuXMgSi₂O₈ of these or Sr₃-xEuXMgSi₂O₈ grade receives the exposure of the light of 350 to 415 nm from the 1st emitter. The above-mentioned fluorescent substance generated the light of high reinforcement more overwhelmingly than Y₃aluminum5O₁₂:Ce of BaMgAl₁₀O₁₇:Eu of general blue luminescence, or yellow luminescence.

[0007]

[Embodiment of the Invention] This invention is luminescence equipment which combined the 2nd emitter which is the 1st emitter and fluorescent substance which generate the light of 350 to 415 nm, and the 2nd emitter is characterized by coming to contain the fluorescent substance which has the crystal phase which has the chemical composition of the following general formula [1].

[0008]

[Formula 3] $M1aEubM2cM3dOe$ [1]

at least a kind of element chosen from the group which M1 becomes from Ba, Sr, and calcium here — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M1 were chosen from the group which consists of Ba, Sr, and calcium especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M1, it is desirable to contain Ba and/or Sr and it is desirable especially to contain Ba. Moreover, when it contains Ba, as for the mole ratio of Ba to Sr from points, such as the ease of carrying out of luminescence wavelength accommodation, and luminescence reinforcement, it is desirable to carry out to 0.05 or more preferably, in this case, the amount of Sr — 0 — you may be (in this case, the above-mentioned mole ratio becomes infinite) — Sr is also made to contain preferably and the above-mentioned mole ratio is usually made or less into 100.

[0009] at least a kind of element chosen from the group which M2 becomes from Mg and Zn in said general formula [1] — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M2 were chosen from the group which consists of Mg and Zn especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M2, it is desirable especially to contain Mg.

[0010] at least a kind of element chosen from the group which M3 becomes from Si and germanium in said general formula [1] — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M3 were chosen from the group which consists of Si and germanium especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M3, it is desirable especially to contain Si.

[0011] as the metallic element in M1, M2, and M3 — ten-mol% — an upper limit — desirable — five-mol% — an upper limit — metallic elements other than the above can be made to contain during a crystal by making three-mol% into an upper limit still more preferably. In this case, respectively, although there is especially no constraint in that metallic element, if the same valence as Ba, Mg, and Si, i.e., divalent, divalent, and a tetravalent metallic element are made to contain, since it will be easy to hold the crystal structure, it is desirable in order. In the semantics which helps crystallization of the multiple oxide by the diffusion in a solid-state at the time of baking of Eu^{2+} which is divalent and tetravalent a metallic element and an emission center, little installation of the metallic elements, such as univalent, trivalent, pentavalence, or 6 **, may be carried out. If an example is given, a part of Ba^{2+} in a $Ba_3MgSi_2O_8:Eu$ fluorescent substance or Mg^{2+} can be permuted holding a charge compensation effect by Li^+ and Ga^{2+} of equimolar.

[0012] In said general formula [1], a, b, c, d, and e express the mole ratio of the metallic element which corresponds to M1 in order, respectively, the mole ratio of an europium atom, the mole ratio of the metallic element applicable to M2, the mole ratio of the metallic element applicable to M3, and the mole ratio of an oxygen atom. Although the value of a+b, and c, d and e is set in general to 3, 1, 2, and 8 at order, even if some cation deficits and oxygen deficiencies have arisen somewhat, it usually has the tolerance which includes the above-mentioned value for the reasons of there being no big effect in the fluorescence engine performance.

[0013] Although a is a number with which are satisfied of $2.5 \leq a \leq 3.3$, it is 2.9 or more numbers still more preferably, and is 3.1 or less number still more preferably 3.2 or less 2.8 or more more preferably 2.7 or more. Although b is a number with which are satisfied of $0.0001 \leq b \leq 1.0$, it is 0.003 or more numbers more preferably, and is 0.1 or less number especially preferably 0.15 or less still more preferably 0.3 or less more preferably 0.5 or less 0.001 or more preferably. There is an inclination for luminescence reinforcement to become [the content of emission center ion] small under in said range, and, on the other hand, there is an inclination for luminescence reinforcement to decrease too, according to the phenomenon in which said excess of the range is also called concentration quenching. Moreover, it is desirable to fill $2.7 \leq a+b \leq 3.3$ in that a crystal phase with few crystal defects is obtained, and luminescence reinforcement becomes high. Although c is a number with which are satisfied of $0.9 \leq c \leq 1.1$, it is 0.95 or more numbers still more preferably, and is 1.05 or less number still more preferably 1.07 or less preferably 0.93 or more. Although d is a number with which are satisfied of $1.8 \leq d \leq 2.2$, it is 1.9 or more numbers still more preferably, and is 2.1 or less number still more preferably 2.15 or less preferably 1.85 or more. Although e is a number with which are satisfied of $7.2 \leq e \leq 8.8$, it is 7.8 or more numbers most preferably 7.6 or more still more preferably 7.4 or more, and is 8.2 or less number most preferably 8.4 or less still more preferably 8.6 or less.

[0014] The typical crystal structure of the fluorescent substance used by this invention is $2OBa_3MgSi_8$ structure, $2OSr_3MgSi_8$ structure, or $2Ocalcium_3MgSi_8$ structure. $2Ocalcium_3MgSi_8$ structure is usually called merwinite structure. $2OBa_3MgSi_8$ structure and $2OSr_3MgSi_8$ structure are strictly considered to be the similar structure instead of merwinite structure. $2OBa_3MgSi_8$ structure and $2OSr_3MgSi_8$ structure are orthorhombic system, and those lattice constants are usually $a = 5.5A$, $b = 9.8A$, $c = 7.6A$ and $a = 5.4A$, $b = 9.6A$, and about $c = 7.2A$. $2Ocalcium_3MgSi_8$ structures are monoclinic system and space groups $P2_1/a$, and lattice constants are usually $a = 13.254A$, $b = 5.293A$, and about $c = 9.328A$. The X diffraction pattern of Ba_3MgSi_8 2O8, Sr_3MgSi_8 2O8, and $calcium_3MgSi_8$ 2O8 is shown in drawing 1, and 2 and 3, respectively (powder X diffraction database). In these crystal structures, since the presentation range where each dissolves each other is wide, as for the divalent metal of Ba, Sr, calcium, and others, it is considered that structure is near. The crystal phase of the fluorescent substance used by this invention corresponds to the thing which made Eu^{2+} permute as an activator to the matter which usually has $2OBa_3MgSi_8$ structure, $2OSr_3MgSi_8$ structure, or $2Ocalcium_3MgSi_8$ structure.

[0015] The fluorescent substance used by this invention is excited by the light of 350 to 415 nm from the 1st emitter, and generates the light. The above-mentioned fluorescent substance generates the light of very strong luminescence reinforcement by excitation of the light of 350 to 415 nm. The fluorescent substance used by this invention is M1 as shown in said general formula [1]. Source, M2 source and M3 In the compound of a source, and a list, the source compound of element (Eu) of emission center ion After grinding using dry mills, such as a hammer mill, a roll mill, a ball mill, and a jet mill, The dry process pulverized using a dry mill after mixing with mixers, such as a ribbon blender, a V type blender, and a Henschel mixer, or mixing, or [or / grinding and mixing by adding these compounds into media, such as water using wet grinding mills, such as a medium stirring type grinder,] -- or After a dry mill grinds these

compounds, it can manufacture by heat-treating and calcinating the prepared grinding mixture with the wet method which dries the slurry prepared by mixing [be / it / under / media /, such as water, / adding] by spray drying etc. [0016] In these grinding mixing methods, it sets to the source compound of an element of emission center ion especially. It is desirable to use a liquid medium, since it is necessary to make the whole mix and distribute a little compound to homogeneity. A latter wet method is desirable also from the field where uniform mixing is obtained by the whole in other source compounds of an element. Moreover, as a heating approach In heat-resistant containers, such as an alumina, crucible made from a quartz, and a tray, 800–1600 degrees C is usually 1000–1400 degrees C in temperature preferably. It is made [heat / independent or / of gases, such as atmospheric air, oxygen, a carbon monoxide, a carbon dioxide, nitrogen, hydrogen, and an argon, / for 10 minutes to 24 hours / under a mixed ambient atmosphere]. In addition, washing, desiccation, classification processing, etc. are made after heat-treatment if needed.

[0017] In addition, an ambient atmosphere required in order to acquire the ionic state (valence) which the element of emission center ion contributes to luminescence as said heating ambient atmosphere is chosen. In divalent Eu in this invention etc., the bottom of the neutrality of a carbon monoxide, nitrogen, hydrogen, an argon, etc. or reducing atmosphere is desirable, but it is possible if the bottom of oxidizing atmospheres, such as atmospheric air and oxygen, also chooses even conditions. In the compound of M1 source, M2 source, and M3 source, and a list here moreover, as a source compound of an element of emission center ion In M1, M2 and M3, and a list, each oxide of the element of emission center ion, a hydroxide, A carbonate, a nitrate, a sulfate, an oxalate, carboxylate, a halogenide, etc. are mentioned, and it is chosen in consideration of un-generating nature, such as reactivity to the multiple oxide out of these and NOx at the time of baking, and SOx, etc.

[0018] If those source compounds of M1 are illustrated concretely, about said Ba, Sr, and calcium which are made desirable to a metallic element M1 as a source compound of Ba BaO, Ba(OH)2.8H2O, BaCO3, Ba (NO3)2 and BaSO4, Ba(OCO) 2.2H2O, Ba (OCOCH3)2 and BaCl2 grade again as a source compound of Sr SrO, Sr(OH)2.8H2O, SrCO3, Sr (NO3)2 and SrSO4, Sr(OCO) 2andH2O, Sr(OCOCH3) 2.0.5H2O and SrCl2 grade again as a source compound of calcium CaO and calcium (OH)2, CaCO3, calcium(NO3) 2.4H2O, CaSO4.2H2O, calcium(OCO) 2andH2O, calcium(OCOCH3) 2andH2O, and CaCl2 grade are mentioned, respectively.

[0019] If those source compounds of M2 are illustrated concretely, about said Mg and Zn which are made desirable to a metallic element M2 moreover, as a source compound of Mg MgO and Mg (OH)2, MgCO3, Mg(OH)2 and 3MgCO3.3H2O, Mg(NO3)2.6H2O, MgSO4, Mg(OCO) 2.2H2O, Mg(OCOCH3) 2.4H2O, and MgCl2 grade again as a source compound of Zn ZnO and Zn (OH)2, ZnCO3, Zn (NO3)2, Zn (OCO)2, Zn (OCOCH3)2, and ZnCl2 grade are mentioned, respectively.

[0020] moreover — if those source compounds of M3 are concretely illustrated about said Si and germanium which are made desirable to a metallic element M3 — the source compound of Si — carrying out — SiO2, H4SiO4, and Si(OCOCH3) 4 grade — moreover — the source compound of germanium — carrying out — GeO2 and germanium (OH)4, germanium (OCOCH3)4, and GeCl4 grade are mentioned, respectively. Furthermore, about said Eu made desirable as an element of emission center ion, if the source compound of an element is illustrated concretely, Eu 2O3, Eu2(SO4) 3, Eu2(OCO) 6, EuCl2, and EuCl3 grade will be mentioned.

[0021] In this invention, the 1st emitter which irradiates light at said fluorescent substance generates the light of 350 to 415 nm wavelength. The emitter which generates the light which has peak wavelength is preferably used for the range of 350 to 415 nm wavelength. As an example of the 1st emitter, light emitting diode (LED) or laser diode (LD) can be mentioned. Laser diode is desirable at the point which can control especially power consumption. Moreover, the GaN systems LED and LD which used the GaN system compound semiconductor are desirable. the GaN systems LED and LD are compared with the SiC system LED which emits the light of this field, a radiant power output and external quantum efficiency are alike and large, and it is because very bright luminescence is dramatically obtained with low power by combining with said fluorescent substance. For example, a GaN system usually has the luminescence reinforcement of 100 times or more of a SiC system to a 20mA current load. In the GaN systems LED and LD, what has the AlXGaYN luminous layer, the GaN luminous layer, or the InXGaYN luminous layer is desirable. In the GaN system LED, since what has an InXGaYN luminous layer in them is dramatically strong as for luminescence reinforcement, it is especially desirable, and in the GaN system LD, since luminescence reinforcement has the dramatically strong thing of the multiplex quantum well structure of an InXGaYN layer and a GaN layer, it is especially desirable. In addition, in the above, the value of X+Y is usually a value of the range of 0.8–1.2. In the GaN system LED, when the thing and what has a nothing dopant that doped Zn and Si to these luminous layers adjust a luminescence property, it is desirable. As for luminous efficiency, what has the hetero structure which used these luminous layers, p layers, n layers, the electrode, and the substrate as the basic component, and made the luminous layer sandwiches in the AlXGaYN layer of n mold and p mold, the GaN layer, or the InXGaYN layer is high, it is desirable, and what made hetero structure quantum well structure further has still higher luminous efficiency, and, as for the GaN system LED, is more desirable.

[0022] In this invention, since the luminous efficiency of the whole luminescence equipment will be raised, it is especially desirable to use the emitter of a surface-emitting type, especially surface-emitting type GaN system laser diode as the 1st emitter. The emitter of a surface-emitting type is an emitter which has luminescence strong against the membranous direction of a field, and luminescence of the direction of a field can be made stronger than the direction of an edge of a luminous layer in surface-emitting type GaN system laser diode by controlling crystal growth, such as a luminous layer, and devising a reflecting layer etc. well. Since exposure area can be dramatically enlarged with the same quantity of light and exposure effectiveness can be improved when irradiating the light at the fluorescent substance of the 2nd emitter, as a result of the luminescence cross section per amount of unit luminescence being able to take greatly compared with the type which emits light from the edge of a luminous layer by using the thing of a surface-emitting type, stronger luminescence can be obtained from the fluorescent substance which is the 2nd emitter.

[0023] When using the thing of a surface-emitting type as the 1st emitter, it is desirable to make the 2nd emitter into the shape of film. Consequently, since the cross section is large enough, if the 2nd emitter is made into the shape of film in the direction of the cross section, since the exposure cross section to the fluorescent substance from the 1st emitter will become large per fluorescent substance unit quantity, the light from the emitter of a surface-emitting type can enlarge reinforcement of luminescence from a fluorescent substance more.

[0024] Moreover, when using the thing of a surface-emitting type as the 1st emitter and using a film-like thing as the 2nd emitter, it is desirable to consider as ***** which contacts the direct film-like 2nd emitter to the luminescence side of the 1st emitter. contact here — the 1st emitter — ** — it says building the condition of having touched

exactly without the 2nd emitter's minding air and a gas. Consequently, since quantity of light loss that it is reflected by the film surface of the 2nd emitter, and the light from the 1st emitter oozes outside is avoidable, luminous efficiency of the whole equipment can be improved.

[0025] The typical perspective view showing the physical relationship of the 1st emitter and the 2nd emitter in an example of the luminescence equipment of this invention is shown in drawing 4. The film-like 2nd emitter with which one in drawing 4 has said fluorescent substance, and 2 express the surface-emitting type GaN system LD as the 1st emitter, and 3 expresses a substrate. In order to build the condition of having contacted mutually, separate ** is built with LD2 and the 2nd emitter 1, respectively, those fields may be contacted with the means of adhesives or others, and the 2nd ***** may be made to produce on the luminescence side of LD2 (molding). LD2 and the 2nd emitter 1 can be made into the condition of having contacted, these results.

[0026] Although the light from the 1st emitter and the light from the 2nd emitter are usually suitable in all directions, since the part will be reflected when light comes out of resin if the powder of the fluorescent substance of the 2nd emitter is distributed in resin, the sense of light can be arranged to some extent. Therefore, since light can be guided to some extent to the efficient sense, it is desirable to use what distributed the powder of said fluorescent substance into resin as the 2nd emitter. Moreover, if a fluorescent substance is distributed in resin, since a total exposure area to the 2nd emitter of the light from the 1st emitter will become large, it also has the advantage that luminescence reinforcement from the 2nd emitter can be enlarged. In this case, as resin which can be used, although various kinds of things, such as an epoxy resin, polyvinyl system resin, polyethylene system resin, a polypropylene resin, and polyester system resin, are mentioned, the dispersibility of fluorescent substance powder is an epoxy resin preferably at a good point. When distributing the powder of the 2nd emitter in resin, the weight ratio of the whole powder concerned of the 2nd emitter and resin powder is 10 - 95% usually 30 - 80% still more preferably 20 to 90% preferably. When there are too many fluorescent substances, luminous efficiency may fall by condensation of powder, and when too few, luminous efficiency may fall shortly for the absorption of light by resin, or dispersion.

[0027] Come to consist of the luminescence equipment of this invention said fluorescent substance as a wavelength conversion ingredient, and a light emitting device which generates the light of 350 to 415 nm, and a fluorescent substance absorbs the light of 350 to 415 nm which a light emitting device emits in the first half. It is not based on an operating environment, but color rendering properties are good, and are luminescence equipment which can be made to generate the light of high intensity, and it is suitable for the light source of lighting systems, such as image display devices, such as sources of luminescence, such as the back light light source and a signal, and a color liquid crystal display, and field luminescence, etc.

[0028] When the luminescence equipment of this invention is explained based on a drawing, drawing 5 It is the typical sectional view showing one example of the luminescence equipment which has the 1st emitter (350-415nm emitter) and 2nd emitter. 4 — for an inner lead and 7, as for the fluorescent substance content resin section as the 2nd emitter, and 9, the 1st emitter (emitter of 350 to 415 nm) and 8 are [luminescence equipment and 5 / a mounting lead and 6 / a conductive wire and 10] mould members.

[0029] As the luminescence equipment which is an example of this invention is shown in drawing 5, the gestalt of a common shell mold in the up cup of nothing and the mounting lead 5 The 1st emitter (350-415nm emitter) 7 which consists of GaN system light emitting diode etc. Moreover binders, such as an epoxy resin and acrylic resin, are made to mix and distribute a fluorescent substance, and it is fixed by slushing in a cup by being covered with the fluorescent substance content resin section 8 formed as the 2nd emitter. On the other hand, it has flowed through an emitter 7, the mounting lead 5, and the 1st emitter 7 and inner lead 6 with the conductive wire 9, respectively, and these whole is covered with the mould member 10 by an epoxy resin etc., and it comes to protect them. [1st]

[0030] Moreover, the field luminescence lighting system 98 incorporating this light emitting device 1 It is a power source, a circuit, etc. for actuation of much luminescence equipments 91 of a light emitting device 91 on the outside to the base of the maintenance case 910 of the rectangle which made the inner surface light impermeability nature, such as a white smooth side, as shown in drawing 9 (it does not illustrate.). It prepares and arranges and comes to fix to the part equivalent to the covering device of the maintenance case 910 the diffusion plates 99, such as an acrylic board made into opalescence, for equalization of luminescence.

[0031] And drive the field luminescence lighting system 98 and the light of 350 to 415 nm is made to emit light by impressing an electrical potential difference to the 1st emitter of a light emitting device 91. Said fluorescent substance in the fluorescent substance content resin section as the 2nd emitter absorbs a part of the luminescence. Light is emitted in the light and, on the other hand, high luminescence of color rendering properties is obtained with color mixture with the blue glow which was not absorbed by the fluorescent substance, this light will penetrate the diffusion plate 99, outgoing radiation will be carried out to the drawing upper part, and the illumination light of uniform brightness will be obtained in the 99th page of the diffusion plate of the maintenance case 910.

[0032]

[Example] Hereafter, although an example explains this invention still more concretely, this invention is not limited to the following examples, unless the summary is exceeded. In addition, relative intensity shows the magnitude of the luminescence reinforcement when setting luminescence reinforcement of the fluorescent substance of the example 1 of a comparison to 100.

As a source compound of example 1M1, as 3; 0.0553 mol of BaCO(s), and a source compound of M2 Basic magnesium carbonate (mol of Mg a number 0.0186 mols), 2O3; 0.00018 mol of Eu(s) as a source compound of an element of emission center ion in 2; 0.0372 mol of SiO(s), and a list as a source compound of M3 and with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1200 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing.

[0033] The X diffraction pattern of fluorescent substance Ba₂.98Eu_{0.02}MgSi 2O8 obtained by drawing 6 is shown. It turns out that the peak pattern of drawing 6 is in agreement with it of Ba₃MgSi 2O8 of drawing 1 in crystal structure. The emission spectrum when exciting this fluorescent substance to drawing 7 by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode is shown. The wavelength and relative intensity of the luminescence peak are shown in a table -1.

[0034] The outside which used 3; 0.0553 mol of SrCO(s) as a source compound of example 2M1 manufactured the fluorescent substance like the example 1. The X diffraction pattern of this fluorescent substance Sr₂.98Eu_{0.02}MgSi

2O8 is shown in drawing 8. It turns out that the peak pattern of drawing 8 is in agreement with it of Sr3MgSi 2O8 of drawing 2 in crystal structure. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0035] The outside which used 3; 0.0442 mol of BaCO(s), 3; 0.0084 mol of CaCO(s), and 3; 0.0028 mol of MnCO(s) as a source compound of example 3M1 manufactured the fluorescent substance like the example 1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0036] The outside which used basic zinc carbonate (mol of Zn a number 0.0186 mols) as a source compound of example 4M2 manufactured the fluorescent substance like the example 1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0037] The fluorescent substance was manufactured like the example 1 except having changed the number of mols of Eu 2O3 which is the source compound of an element of example 5 emission-center ion with 0.000074 mols, and having changed the number of mols of BaCO3 with 0.0556 mols as a source compound of M1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0038] The fluorescent substance was manufactured like the example 1 except having changed example 6 burning temperature with 1300 degrees C. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode were shown in a table -1.

[0039] Fluorescent substance Ba2.95Eu0.05MgSi 2O8 was manufactured like the example 1 except having changed the loadings of example 7BaCO3 into 0.0549 mols, and having changed the loadings of Eu 2O3 into 0.00047 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 438nm, and relative intensity was 299.

[0040] Fluorescent substance Ba2.9Eu0.1MgSi 2O8 was manufactured like the example 1 except having changed the loadings of example 8BaCO3 into 0.0539 mols, and having changed the loadings of Eu 2O3 into 0.00093 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440nm, and relative intensity was 320.

[0041] Fluorescent substance Ba2.85Eu0.15MgSi 2O8 was manufactured like the example 1 except having changed the loadings of example 9BaCO3 into 0.0530 mols, and having changed the loadings of Eu 2O3 into 0.00140 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440, and relative intensity was 261.

[0042] Fluorescent substance Ba2.8Eu0.2MgSi 2O8 was manufactured like the example 1 except having changed the loadings of example 10BaCO3 into 0.0521 mols, and having changed the loadings of Eu 2O3 into 0.00186 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440, and relative intensity was 199.

[0043] Fluorescent substance Ba2.7Eu0.3MgSi 2O8 was manufactured like the example 1 except having changed the loadings of example 11BaCO3 into 0.0502 mols, and having changed the loadings of Eu 2O3 into 0.00279 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 441, and relative intensity was 113.

[0044] 3; 0.0103 mol of example of comparison 1BaCO(s), basic magnesium carbonate (mol of Mg a number 0.0103 mols), 2O3; 0.00057 mol of Eu(s) as a source compound of an element of emission center ion in 2O3; 0.0570 mol of gamma-aluminum, and a list and with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1500 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance (Ba0.9</SUB> Eu0.1MgAl 10O17) of blue luminescence was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing. The emission spectrum when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was shown in drawing 7, and the engine performance of the blue luminescence fluorescent substance of an example 1 and the example 1 of a comparison was compared with it. The wavelength and relative intensity of the luminescence peak were shown in a table -1. A certain thing understands the luminescence reinforcement of the fluorescent substance of the example 1 by 400nm excitation also 2.8 times of that of the fluorescent substance of the example 1 of a comparison.

[0045] 2; 0.00048 mol of CeO(s) as a source compound of an element of emission center ion in 2Y2O3; 0.0238 mols of examples of a comparison, 2O3; 0.0400 mol of gamma-aluminum, and a list with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1500 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance (Y2.98Ce0.03aluminum 5O12) of yellow luminescence was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing. The wavelength and relative intensity of the luminescence peak were shown in a table -1. A certain thing understands the luminescence reinforcement of the fluorescent substance of the example 1 by 400nm excitation also 250 times of that of the fluorescent substance of the example 2 of a comparison.

[0046]

[A table 1]

表 - 1

実施例又は比較例	蛍光体の化学組成	発光波長 (nm)	相対発光強度
実施例1	$\text{Ba}_{1.0}\text{Eu}_{0.9}\text{MgSi}_2\text{O}_8$	434	277
実施例2	$\text{Sr}_{1.0}\text{Eu}_{0.9}\text{MgSi}_2\text{O}_8$	455	238
実施例3	$\text{Ba}_{1.0}\text{Eu}_{0.9}\text{Ca}_{0.1}\text{Mn}_{0.1}\text{MgSi}_2\text{O}_8$	440	227
実施例4	$\text{Ba}_{1.0}\text{Eu}_{0.9}\text{ZnSi}_2\text{O}_8$	436	255
実施例5	$\text{Ba}_{1.0}\text{Eu}_{0.9}\text{MgSi}_2\text{O}_8$	435	235
実施例6	$\text{Ba}_{1.0}\text{Eu}_{0.9}\text{MgSi}_2\text{O}_8$	434	277
比較例1	$\text{Ba}_{0.9}\text{Eu}_{0.1}\text{MgAl}_2\text{O}_7$	448	100
比較例2	$\text{Y}_{2.0}\text{Ce}_{0.9}\text{Al}_2\text{O}_7$	561	1.1

[0047]

[Effect of the Invention] According to this invention, the high luminescence equipment of luminescence reinforcement can be offered.

[Translation done.]

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TECHNICAL FIELD

[Field of the Invention] The 1st emitter to which this invention emits light in the light of a light field from ultraviolet radiation by the power source in detail about luminescence equipment, By combining the 2nd emitter which has the fluorescent substance with which the parent compound which absorbs the light which is in a light field from the ultraviolet radiation, and emits the light of long wavelength contains emission center ion and which carries out a wavelength conversion ingredient It is not based on an operating environment, but color rendering properties are good and are related with the luminescence equipment which can be made to generate luminescence of high intensity.

[Translation done.]

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PRIOR ART

[Description of the Prior Art] The luminescence equipment which carried out color conversion of the luminescent color of LED or LD with the fluorescent substance with blue, red, and green color mixture in order to generate various colors of white and others with color rendering properties uniform and sufficient is proposed. For example, by JP,49-1221,B, the beam of the laser which emits the radiation beam of the wavelength of 300 to 530 nm is made to irradiate a phosphor ($Y_3-x-yCexGdyM5-zGazO_{12}$ (for Y to express La and for Y, Lu, or M to express aluminum, aluminum-In, or aluminum-Sc.)), and the approach of making this emit light and forming a display is shown. Moreover, the luminescence equipment of white luminescence which consists of recent years combining the gallium nitride (GaN) systems LED and LD with high luminous efficiency and the fluorescent substance as a wavelength conversion ingredient which attract attention as a semi-conductor light emitting device of blue luminescence is proposed as a source of luminescence of an image display device or a lighting system taking advantage of the description that power consumption is small and it is long lasting. The luminescence equipment characterized by using LED or LD chip of this nitride system semi-conductor, and actually using an yttrium aluminum garnet system as a fluorescent substance in JP,10-242513,A is shown.

[0003] However, despite former, it could not say that the known luminescence equipment which combined the yttrium aluminum garnet system fluorescent substance as shown in JP,10-242513,A as the 2nd emitter was enough as luminescence reinforcement to the 1st emitter, such as LED, but the further amelioration was called for as sources of luminescence, such as a display, and the back light light source, a signal.

[Translation done.]

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EFFECT OF THE INVENTION

[Effect of the Invention] According to this invention, the high luminescence equipment of luminescence reinforcement can be offered.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] This invention is made in view of the above-mentioned conventional technique that the very high luminescence equipment of luminescence reinforcement should be developed, therefore this invention aims at offering obtaining double emitter mold luminescence equipment with very high luminescence reinforcement while it is easy to manufacture.

[Translation done.]

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MEANS

[Means for Solving the Problem] In the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned as a result of inquiring wholeheartedly that this invention person should solve said technical problem When the fluorescent substance containing the crystal phase which has the chemical composition of the following specification as the 2nd emitter of the above was used, said fluorescent substance received the exposure of the light near 350-415nm, it found out that said object could be attained as a result of causing luminescence of the light by high reinforcement, and this invention was reached. That is, let the luminescence equipment characterized by this invention coming to contain the fluorescent substance which has the crystal phase in which said 2nd emitter has the chemical composition of a general formula [1] in the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned be the summary.

[0006]

[Formula 2] $M1aEubM2cM3dOe$ [1]

The included metallic element is expressed. however, at least a kind of element chosen from the group which M1 becomes from Ba, Sr, and calcium — the sum total — more than 90mol% — M2 The included metallic element is expressed. at least a kind of element chosen from the group which consists of Mg and Zn — the sum total — more than 90mol% — M3 The included metallic element is expressed. at least a kind of element chosen from the group which consists of Si and germanium — the sum total — more than 90mol% — the number with which a is satisfied of $2.5 \leq a \leq 3.3$, the number with which b is satisfied of $0.0001 \leq b \leq 1.0$, the number with which c is satisfied of $0.9 \leq c \leq 1.1$, the number with which d is satisfied of $1.8 \leq d \leq 2.2$, and e are numbers with which are satisfied of $7.2 \leq e \leq 8.8$.

In addition, the crystal phase of $Ba_3MgSi_2O_8$ and $Sr_3MgSi_2O_8$ grade itself is known, and it is also known that Ba of these and Sr will be permuted by other divalent metal elements, such as Eu^{2+} , and will get. This invention is based on having found out generating light with notably high reinforcement rather than other fluorescent substances, if the fluorescent substance which has the crystal phase which has the chemical composition of said general formula [1] including the chemical composition of $Ba_3-xEuXMgSi_2O_8$ of these or $Sr_3-xEuXMgSi_2O_8$ grade receives the exposure of the light of 350 to 415 nm from the 1st emitter. The above-mentioned fluorescent substance generated the light of high reinforcement more overwhelmingly than $Y_3Aluminum_5O_{12}:Ce$ of $BaMgAl_{10}O_{17}:Eu$ of general blue luminescence, or yellow luminescence.

[0007]

[Embodiment of the Invention] This invention is luminescence equipment which combined the 2nd emitter which is the 1st emitter and fluorescent substance which generate the light of 350 to 415 nm, and the 2nd emitter is characterized by coming to contain the fluorescent substance which has the crystal phase which has the chemical composition of the following general formula [1].

[0008]

[Formula 3] $M1aEubM2cM3dOe$ [1]

at least a kind of element chosen from the group which M1 becomes from Ba, Sr, and calcium here — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M1 were chosen from the group which consists of Ba, Sr, and calcium especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M1, it is desirable to contain Ba and/or Sr and it is desirable especially to contain Ba. Moreover, when it contains Ba, as for the mole ratio of Ba to Sr from points, such as the ease of carrying out of luminescence wavelength accommodation, and luminescence reinforcement, it is desirable to carry out to 0.05 or more preferably. in this case, the amount of Sr — 0 — you may be (in this case, the above-mentioned mole ratio becomes infinite) — Sr is also made to contain preferably and the above-mentioned mole ratio is usually made or less into 100.

[0009] at least a kind of element chosen from the group which M2 becomes from Mg and Zn in said general formula [1] — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M2 were chosen from the group which consists of Mg and Zn especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M2, it is desirable especially to contain Mg.

[0010] at least a kind of element chosen from the group which M3 becomes from Si and germanium in said general formula [1] — the sum total — more than 90mol% — desirable — more than 95mol% — the included metallic element is expressed. The thing for which all the elements applicable to M3 were chosen from the group which consists of Si and germanium especially and which is a kind at least is desirable. Consequently, bigger luminescence reinforcement can be obtained. As M3, it is desirable especially to contain Si.

[0011] as the metallic element in M1, M2, and M3 — ten-mol% — an upper limit — desirable — five-mol% — an upper limit — metallic elements other than the above can be made to contain during a crystal by making three-mol% into an upper limit still more preferably In this case, respectively, although there is especially no constraint in that metallic element, if the same valence as Ba, Mg, and Si, i.e., divalent, divalent, and a tetravalent metallic element are made to contain, since it will be easy to hold the crystal structure, it is desirable in order. In the semantics which helps crystallization of the multiple oxide by the diffusion in a solid-state at the time of baking of Eu^{2+} which is divalent and tetravalent a metallic element and an emission center, little installation of the metallic elements, such as univalent,

trivalent, pentavalence, or 6 **, may be carried out. If an example is given, a part of Ba²⁺ in a Ba₃MgSi₂O₈:Eu fluorescent substance or Mg²⁺ can be permuted holding a charge compensation effect by Li⁺ and Ga²⁺ of equimolar. [0012] In said general formula [1], a, b, c, d, and e express the mole ratio of the metallic element which corresponds to M1 in order, respectively, the mole ratio of an europium atom, the mole ratio of the metallic element applicable to M2, the mole ratio of the metallic element applicable to M3, and the mole ratio of an oxygen atom. Although the value of a+b, and c, d and e is set in general to 3, 1, 2, and 8 at order, even if some cation deficits and oxygen deficiencies have arisen somewhat, it usually has the tolerance which includes the above-mentioned value for the reasons of there being no big effect in the fluorescence engine performance.

[0013] Although a is a number with which are satisfied of $2.5 \leq a \leq 3.3$, it is 2.9 or more numbers still more preferably, and is 3.1 or less number still more preferably 3.2 or less 2.8 or more more preferably 2.7 or more. Although b is a number with which are satisfied of $0.0001 \leq b \leq 1.0$, it is 0.003 or more numbers more preferably, and is 0.1 or less number especially preferably 0.15 or less still more preferably 0.3 or less more preferably 0.5 or less 0.001 or more preferably. There is an inclination for luminescence reinforcement to become [the content of emission center ion] small under in said range, and, on the other hand, there is an inclination for luminescence reinforcement to decrease too, according to the phenomenon in which said excess of the range is also called concentration quenching. Moreover, it is desirable to fill $2.7 \leq a+b \leq 3.3$ in that a crystal phase with few crystal defects is obtained, and luminescence reinforcement becomes high. Although c is a number with which are satisfied of $0.9 \leq c \leq 1.1$, it is 0.95 or more numbers still more preferably, and is 1.05 or less number still more preferably 1.07 or less preferably 0.93 or more. Although d is a number with which are satisfied of $1.8 \leq d \leq 2.2$, it is 1.9 or more numbers still more preferably, and is 2.1 or less number still more preferably 2.15 or less preferably 1.85 or more. Although e is a number with which are satisfied of $7.2 \leq e \leq 8.8$, it is 7.8 or more numbers most preferably 7.6 or more still more preferably 7.4 or more, and is 8.2 or less number most preferably 8.4 or less still more preferably 8.6 or less.

[0014] The typical crystal structure of the fluorescent substance used by this invention is 2OBa₃MgSi₈ structure, 2OSr₃MgSi₈ structure, or 2Ocalcium₃MgSi₈ structure. 2Ocalcium₃MgSi₈ structure is usually called merwinite structure. 2OBa₃MgSi₈ structure and 2OSr₃MgSi₈ structure are strictly considered to be the similar structure instead of merwinite structure. 2OBa₃MgSi₈ structure and 2OSr₃MgSi₈ structure are orthorhombic system, and those lattice constants are usually a= 5.5Å of *****, b= 9.8Å, c= 7.6Å and a= 5.4Å, b= 9.6Å, and about c= 7.2Å. 2Ocalcium₃MgSi₈ structures are monoclinic system and space groups P2₁/a, and lattice constants are usually a= 13.254Å, b= 5.293Å, and about c= 9.328Å. The X diffraction pattern of Ba₃MgSi₂O₈, Sr₃MgSi₂O₈, and calcium₃MgSi₂O₈ is shown in drawing 1, and 2 and 3, respectively (powder X diffraction database). In these crystal structures, since the presentation range where each dissolves each other is wide, as for the divalent metal of Ba, Sr, calcium, and others, it is considered that structure is near. The crystal phase of the fluorescent substance used by this invention corresponds to the thing which made Eu²⁺ permute as an activator to the matter which usually has 2OBa₃MgSi₈ structure, 2OSr₃MgSi₈ structure, or 2Ocalcium₃MgSi₈ structure.

[0015] The fluorescent substance used by this invention is excited by the light of 350 to 415 nm from the 1st emitter, and generates the light. The above-mentioned fluorescent substance generates the light of very strong luminescence reinforcement by excitation of the light of 350 to 415 nm. The fluorescent substance used by this invention is M1 as shown in said general formula [1]. Source, M2 source and M3 in the compound of a source, and a list, the source compound of element (Eu) of emission center ion After grinding using dry mills, such as a hammer mill, a roll mill, a ball mill, and a jet mill, The dry process pulverized using a dry mill after mixing with mixers, such as a ribbon blender, a V type blender, and a Henschel mixer, or mixing, or [or / grinding and mixing by adding these compounds into media, such as water using wet grinding mills, such as a medium stirring type grinder,] — or After a dry mill grinds these compounds, it can manufacture by heat-treating and calcinating the prepared grinding mixture with the wet method which dries the slurry prepared by mixing [be / it / under / media /, such as water, / adding] by spray drying etc.

[0016] In these grinding mixing methods, it sets to the source compound of an element of emission center ion especially. It is desirable to use a liquid medium, since it is necessary to make the whole mix and distribute a little compound to homogeneity. A latter wet method is desirable also from the field where uniform mixing is obtained by the whole in other source compounds of an element. Moreover, as a heating approach In heat-resistant containers, such as an alumina, crucible made from a quartz, and a tray, 800–1600 degrees C is usually 1000–1400 degrees C in temperature preferably. It is made [heat / independent or / of gases, such as atmospheric air, oxygen, a carbon monoxide, a carbon dioxide, nitrogen, hydrogen, and an argon, / for 10 minutes to 24 hours / under a mixed ambient atmosphere]. In addition, washing, desiccation, classification processing, etc. are made after heat-treatment if needed.

[0017] In addition, an ambient atmosphere required in order to acquire the ionic state (valence) which the element of emission center ion contributes to luminescence as said heating ambient atmosphere is chosen. In divalent Eu in this invention etc., the bottom of the neutrality of a carbon monoxide, nitrogen, hydrogen, an argon, etc. or reducing atmosphere is desirable, but it is possible if the bottom of oxidizing atmospheres, such as atmospheric air and oxygen, also chooses even conditions. In the compound of M1 source, M2 source, and M3 source, and a list here moreover, as a source compound of an element of emission center ion In M1, M2 and M3, and a list, each oxide of the element of emission center ion, a hydroxide, A carbonate, a nitrate, a sulfate, an oxalate, carboxylate, a halogenide, etc. are mentioned, and it is chosen in consideration of un-generating nature, such as reactivity to the multiple oxide out of these and NO_x at the time of baking, and SO_x, etc.

[0018] If those source compounds of M1 are illustrated concretely, about said Ba, Sr, and calcium which are made desirable to a metallic element M1 as a source compound of Ba BaO, Ba(OH)₂·2.8H₂O, BaCO₃, Ba (NO₃)₂ and BaSO₄, Ba(OCO) 2.2H₂O, Ba (OCOCH₃)₂ and BaCl₂ grade again as a source compound of Sr SrO, Sr(OH)₂·2.8H₂O, SrCO₃, Sr (NO₃)₂ and SrSO₄, Sr(OCO) 2andH₂O, Sr(OCOCH₃) 2.0.5H₂O and SrCl₂ grade again as a source compound of calcium CaO and calcium (OH)₂, CaCO₃, calcium(NO₃) 2.4H₂O, CaSO₄·2H₂O, calcium(OCO) 2andH₂O, calcium(OCOCH₃) 2andH₂O, and CaCl₂ grade are mentioned, respectively.

[0019] If those source compounds of M2 are illustrated concretely, about said Mg and Zn which are made desirable to a metallic element M2 moreover, as a source compound of Mg MgO and Mg (OH)₂, MgCO₃, Mg(OH)₂ and 3MgCO₃·3H₂O, Mg(NO₃)₂·6H₂O, MgSO₄, Mg(OCO) 2.2H₂O, Mg(OCOCH₃) 2.4H₂O, and MgCl₂ grade again as a source compound of Zn ZnO and Zn (OH)₂, ZnCO₃, Zn (NO₃)₂, Zn (OCO)₂, Zn (OCOCH₃)₂, and ZnCl₂ grade are mentioned, respectively.

[0020] moreover — if those source compounds of M3 are concretely illustrated about said Si and germanium which are made desirable to a metallic element M3 — the source compound of Si — carrying out — SiO₂, H₄SiO₄, and Si(OCOCH₃) 4 grade — moreover — the source compound of germanium — carrying out — GeO₂ and germanium

(OH)₄, germanium (OCOCH₃)₄, and GeCl₄ grade are mentioned, respectively. Furthermore, about said Eu made desirable as an element of emission center ion, if the source compound of an element is illustrated concretely, Eu₂O₃, Eu₂(SO₄)₃, Eu₂(OCO)₆, EuCl₂, and EuCl₃ grade will be mentioned.

[0021] In this invention, the 1st emitter which irradiates light at said fluorescent substance generates the light of 350 to 415 nm wavelength. The emitter which generates the light which has peak wavelength is preferably used for the range of 350 to 415 nm wavelength. As an example of the 1st emitter, light emitting diode (LED) or laser diode (LD) can be mentioned. Laser diode is desirable at the point which can control especially power consumption. Moreover, the GaN systems LED and LD which used the GaN system compound semiconductor are desirable. the GaN systems LED and LD are compared with the SiC system LED which emits the light of this field, a radiant power output and external quantum efficiency are alike and large, and it is because very bright luminescence is dramatically obtained with low power by combining with said fluorescent substance. For example, a GaN system usually has the luminescence reinforcement of 100 times or more of a SiC system to a 20mA current load. In the GaN systems LED and LD, what has the AlXGaYN luminous layer, the GaN luminous layer, or the InXGaYN luminous layer is desirable. In the GaN system LED, since what has an InXGaYN luminous layer in them is dramatically strong as for luminescence reinforcement, it is especially desirable, and in the GaN system LD, since luminescence reinforcement has the dramatically strong thing of the multiplex quantum well structure of an InXGaYN layer and a GaN layer, it is especially desirable. In addition, in the above, the value of X+Y is usually a value of the range of 0.8–1.2. In the GaN system LED, when the thing and what has a nothing dopant that doped Zn and Si to these luminous layers adjust a luminescence property, it is desirable. As for luminous efficiency, what has the hetero structure which used these luminous layers, p layers, n layers, the electrode, and the substrate as the basic component, and made the luminous layer sandwiches in the AlXGaYN layer of n mold and p mold, the GaN layer, or the InXGaYN layer is high, it is desirable, and what made hetero structure quantum well structure further has still higher luminous efficiency, and, as for the GaN system LED, is more desirable.

[0022] In this invention, since the luminous efficiency of the whole luminescence equipment will be raised, it is especially desirable to use the emitter of a surface-emitting type, especially surface-emitting type GaN system laser diode as the 1st emitter. The emitter of a surface-emitting type is an emitter which has luminescence strong against the membranous direction of a field, and luminescence of the direction of a field can be made stronger than the direction of an edge of a luminous layer in surface-emitting type GaN system laser diode by controlling crystal growth, such as a luminous layer, and devising a reflecting layer etc. well. Since exposure area can be dramatically enlarged with the same quantity of light and exposure effectiveness can be improved when irradiating the light at the fluorescent substance of the 2nd emitter, as a result of the luminescence cross section per amount of unit luminescence being able to take greatly compared with the type which emits light from the edge of a luminous layer by using the thing of a surface-emitting type, stronger luminescence can be obtained from the fluorescent substance which is the 2nd emitter.

[0023] When using the thing of a surface-emitting type as the 1st emitter, it is desirable to make the 2nd emitter into the shape of film. Consequently, since the cross section is large enough, if the 2nd emitter is made into the shape of film in the direction of the cross section, since the exposure cross section to the fluorescent substance from the 1st emitter will become large per fluorescent substance unit quantity, the light from the emitter of a surface-emitting type can enlarge reinforcement of luminescence from a fluorescent substance more.

[0024] Moreover, when using the thing of a surface-emitting type as the 1st emitter and using a film-like thing as the 2nd emitter, it is desirable to consider as ***** which contacts the direct film-like 2nd emitter to the luminescence side of the 1st emitter. contact here — the 1st emitter — ** — it says building the condition of having touched exactly without the 2nd emitter's minding air and a gas. Consequently, since quantity of light loss that it is reflected by the film surface of the 2nd emitter, and the light from the 1st emitter oozes outside is avoidable, luminous efficiency of the whole equipment can be improved.

[0025] The typical perspective view showing the physical relationship of the 1st emitter and the 2nd emitter in an example of the luminescence equipment of this invention is shown in drawing 4. The film-like 2nd emitter with which one in drawing 4 has said fluorescent substance, and 2 express the surface-emitting type GaN system LD as the 1st emitter, and 3 expresses a substrate. In order to build the condition of having contacted mutually, separate ** is built with LD2 and the 2nd emitter 1, respectively, those fields may be contacted with the means of adhesives or others, and the 2nd ***** may be made to produce on the luminescence side of LD2 (molding). LD2 and the 2nd emitter 1 can be made into the condition of having contacted, these results.

[0026] Although the light from the 1st emitter and the light from the 2nd emitter are usually suitable in all directions, since the part will be reflected when light comes out of resin if the powder of the fluorescent substance of the 2nd emitter is distributed in resin, the sense of light can be arranged to some extent. Therefore, since light can be guided to some extent to the efficient sense, it is desirable to use what distributed the powder of said fluorescent substance into resin as the 2nd emitter. Moreover, if a fluorescent substance is distributed in resin, since a total exposure area to the 2nd emitter of the light from the 1st emitter will become large, it also has the advantage that luminescence reinforcement from the 2nd emitter can be enlarged. In this case, as resin which can be used, although various kinds of things, such as an epoxy resin, polyvinyl system resin, polyethylene system resin, a polypropylene resin, and polyester system resin, are mentioned, the dispersibility of fluorescent substance powder is an epoxy resin preferably at a good point. When distributing the powder of the 2nd emitter in resin, the weight ratio of the whole powder concerned of the 2nd emitter and resin powder is 10 – 95% usually 30 – 80% still more preferably 20 to 90% preferably. When there are too many fluorescent substances, luminous efficiency may fall by condensation of powder, and when too few, luminous efficiency may fall shortly for the absorption of light by resin, or dispersion.

[0027] Come to consist of the luminescence equipment of this invention said fluorescent substance as a wavelength conversion ingredient, and a light emitting device which generates the light of 350 to 415 nm, and a fluorescent substance absorbs the light of 350 to 415 nm which a light emitting device emits in the first half. It is not based on an operating environment, but color rendering properties are good, and are luminescence equipment which can be made to generate the light of high intensity, and it is suitable for the light source of lighting systems, such as image display devices, such as sources of luminescence, such as the back light light source and a signal, and a color liquid crystal display, and field luminescence, etc.

[0028] When the luminescence equipment of this invention is explained based on a drawing, drawing 5 It is the typical sectional view showing one example of the luminescence equipment which has the 1st emitter (350–415nm emitter) and

2nd emitter, 4 -- for an inner lead and 7, as for the fluorescent substance content resin section as the 2nd emitter, and 9, the 1st emitter (emitter of 350 to 415 nm) and 8 are [luminescence equipment and 5 / a mounting lead and 6 / a conductive wire and 10] mould members.

[0029] As the luminescence equipment which is an example of this invention is shown in drawing 5, the gestalt of a common shell mold in the up cup of nothing and the mounting lead 5 The 1st emitter (350-415nm emitter) 7 which consists of GaN system light emitting diode etc. Moreover binders, such as an epoxy resin and acrylic resin, are made to mix and distribute a fluorescent substance, and it is fixed by slushing in a cup by being covered with the fluorescent substance content resin section 8 formed as the 2nd emitter. On the other hand, it has flowed through an emitter 7, the mounting lead 5, and the 1st emitter 7 and inner lead 6 with the conductive wire 9, respectively, and these whole is covered with the mould member 10 by an epoxy resin etc., and it comes to protect them. [1st]

[0030] Moreover, the field luminescence lighting system 98 incorporating this light emitting device 1 It is a power source, a circuit, etc. for actuation of much luminescence equipments 91 of a light emitting device 91 on the outside to the base of the maintenance case 910 of the rectangle which made the inner surface light impermeability nature, such as a white smooth side, as shown in drawing 9 (it does not illustrate.). It prepares and arranges and comes to fix to the part equivalent to the covering device of the maintenance case 910 the diffusion plates 99, such as an acrylic board made into opalescence, for equalization of luminescence.

[0031] And drive the field luminescence lighting system 98 and the light of 350 to 415 nm is made to emit light by impressing an electrical potential difference to the 1st emitter of a light emitting device 91. Said fluorescent substance in the fluorescent substance content resin section as the 2nd emitter absorbs a part of the luminescence. Light is emitted in the light and, on the other hand, high luminescence of color rendering properties is obtained with color mixture with the blue glow which was not absorbed by the fluorescent substance, this light will penetrate the diffusion plate 99, outgoing radiation will be carried out to the drawing upper part, and the illumination light of uniform brightness will be obtained in the 99th page of the diffusion plate of the maintenance case 910.

[Translation done.]

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EXAMPLE

[Example] Hereafter, although an example explains this invention still more concretely, this invention is not limited to the following examples, unless the summary is exceeded. In addition, relative intensity shows the magnitude of the luminescence reinforcement when setting luminescence reinforcement of the fluorescent substance of the example 1 of a comparison to 100.

As a source compound of example 1M1, as 3; 0.0553 mol of BaCO(s), and a source compound of M2 Basic magnesium carbonate (mol of Mg a number 0.0186 mols), 2O3; 0.00018 mol of Eu(s) as a source compound of an element of emission center ion in 2; 0.0372 mol of SiO(s), and a list as a source compound of M3 and with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1200 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing.

[0033] The X diffraction pattern of fluorescent substance Ba_{2.98}Eu_{0.02}MgSi 2O₈ obtained by drawing 6 is shown. It turns out that the peak pattern of drawing 6 is in agreement with it of Ba₃MgSi 2O₈ of drawing 1 in crystal structure. The emission spectrum when exciting this fluorescent substance to drawing 7 by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode is shown. The wavelength and relative intensity of the luminescence peak are shown in a table -1.

[0034] The outside which used 3; 0.0553 mol of SrCO(s) as a source compound of example 2M1 manufactured the fluorescent substance like the example 1. The X diffraction pattern of this fluorescent substance Sr_{2.98}Eu_{0.02}MgSi 2O₈ is shown in drawing 8. It turns out that the peak pattern of drawing 8 is in agreement with it of Sr₃MgSi 2O₈ of drawing 2 in crystal structure. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0035] The outside which used 3; 0.0442 mol of BaCO(s), 3; 0.0084 mol of CaCO(s), and 3; 0.0028 mol of MnCO(s) as a source compound of example 3M1 manufactured the fluorescent substance like the example 1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0036] The outside which used basic zinc carbonate (mol of Zn a number 0.0186 mols) as a source compound of example 4M2 manufactured the fluorescent substance like the example 1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0037] The fluorescent substance was manufactured like the example 1 except having changed the number of mols of Eu 2O₃ which is the source compound of an element of example 5 emission-center ion with 0.000074 mols, and having changed the number of mols of BaCO₃ with 0.0556 mols as a source compound of M1. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode are shown in a table -1.

[0038] The fluorescent substance was manufactured like the example 1 except having changed example 6 burning temperature with 1300 degrees C. The wavelength and relative intensity of a luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode were shown in a table -1.

[0039] Fluorescent substance Ba_{2.95}Eu_{0.05}MgSi 2O₈ was manufactured like the example 1 except having changed the loadings of example 7BaCO₃ into 0.0549 mols, and having changed the loadings of Eu 2O₃ into 0.00047 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 438nm, and relative intensity was 299.

[0040] Fluorescent substance Ba_{2.9}Eu_{0.1}MgSi 2O₈ was manufactured like the example 1 except having changed the loadings of example 8BaCO₃ into 0.0539 mols, and having changed the loadings of Eu 2O₃ into 0.00093 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440nm, and relative intensity was 320.

[0041] Fluorescent substance Ba_{2.85}Eu_{0.15}MgSi 2O₈ was manufactured like the example 1 except having changed the loadings of example 9BaCO₃ into 0.0530 mols, and having changed the loadings of Eu 2O₃ into 0.00140 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440, and relative intensity was 261.

[0042] Fluorescent substance Ba_{2.8}Eu_{0.2}MgSi 2O₈ was manufactured like the example 1 except having changed the loadings of example 10BaCO₃ into 0.0521 mols, and having changed the loadings of Eu 2O₃ into 0.00186 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 440, and relative intensity was 199.

[0043] Fluorescent substance Ba_{2.7}Eu_{0.3}MgSi 2O₈ was manufactured like the example 1 except having changed the loadings of example 11BaCO₃ into 0.0502 mols, and having changed the loadings of Eu 2O₃ into 0.00279 mols. The wavelength of the luminescence peak when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was 441, and relative intensity was 113.

[0044] 3; 0.0103 mol of example of comparison 1BaCO(s), basic magnesium carbonate (mol of Mg a number 0.0103

mols), 2O₃; 0.00057 mol of Eu(s) as a source compound of an element of emission center ion in 2O₃; 0.0570 mol of gamma-aluminum, and a list and with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1500 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance (Ba_{0.9}Eu_{0.1}MgAl₁₀O₁₇) of blue luminescence was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing. The emission spectrum when exciting this fluorescent substance by 400nm which is the dominant wavelength of the ultraviolet radiation field of GaN system light emitting diode was shown in drawing 7, and the engine performance of the blue luminescence fluorescent substance of an example 1 and the example 1 of a comparison was compared with it. The wavelength and relative intensity of the luminescence peak were shown in a table -1. A certain thing understands the luminescence reinforcement of the fluorescent substance of the example 1 by 400nm excitation also 2.8 times of that of the fluorescent substance of the example 1 of a comparison.

[0045] 2; 0.00048 mol of CeO(s) as a source compound of an element of emission center ion in 2Y₂O₃; 0.0238 mols of examples of a comparison, 2O₃; 0.0400 mol of gamma-aluminum, and a list with pure water After grinding and mixing in the container made from an alumina, and the wet ball mill of a bead and passing a nylon mesh after desiccation, the obtained grinding mixture in the crucible made from an alumina It calcinated by heating at 1500 degrees C for 2 hours under the nitrogen gas stream containing 4% of hydrogen, and the fluorescent substance (Y_{2.98}Ce_{0.03}aluminum 5O₁₂) of yellow luminescence was succeedingly manufactured by performing backwashing by water, desiccation, and classification processing. The wavelength and relative intensity of the luminescence peak were shown in a table -1. A certain thing understands the luminescence reinforcement of the fluorescent substance of the example 1 by 400nm excitation also 250 times of that of the fluorescent substance of the example 2 of a comparison.

[0046]

[A table 1]

表 - 1

実施例又は比較例	蛍光体の化学組成	発光ピーク波長 (nm)	相対発光強度
実施例1	Ba _{2.0} Eu _{0.0} MgSi ₂ O ₈	434	277
実施例2	Sr _{2.0} Eu _{0.0} MgSi ₂ O ₈	455	238
実施例3	Ba _{2.0} Eu _{0.0} Ca _{0.4} Mn _{0.1} MgSi ₂ O ₈	440	227
実施例4	Ba _{2.0} Eu _{0.0} ZnSi ₂ O ₈	436	255
実施例5	Ba _{2.0} Eu _{0.0} MgSi ₂ O ₈	435	225
実施例6	Ba _{2.0} Eu _{0.0} MgSi ₂ O ₈	434	277
比較例1	Ba _{0.9} Eu _{0.1} MgAl ₁₀ O ₁₇	448	100
比較例2	Y _{2.98} Ce _{0.03} Al ₅ O ₁₂	561	1.1

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] The X diffraction pattern of Ba₃MgSi₂O₈ (what was converted into X line source Cu Kalpha)

[Drawing 2] The X diffraction pattern of Sr₃MgSi₂O₈ (what was converted into X line source Cu Kalpha)

[Drawing 3] The X diffraction pattern of calcium₃MgSi₂O₈ (what was converted into X line source Cu Kalpha)

[Drawing 4] The typical perspective view showing the physical relationship of the 1st emitter and the 2nd emitter in an example of the luminescence equipment of this invention.

[Drawing 5] The typical sectional view showing one example of the luminescence equipment which has the 1st emitter (350-415nm emitter) and 2nd emitter.

[Drawing 6] The X diffraction pattern of the fluorescent substance of an example 1 (X line source: Cu Kalpha)

[Drawing 7] The emission spectrum when combining the fluorescent substance of the example 1 of this invention, and the example 1 of a comparison with GaN system light emitting diode with a luminescence wavelength of 400nm, respectively.

[Drawing 8] The X diffraction pattern of the fluorescent substance of an example 2 (X line source: Cu Kalpha)

[Drawing 9] The typical sectional view showing an example of the field luminescence lighting system of this invention.

[Description of Notations]

1; the 2nd emitter

2; surface-emitting type GaN system LED

3; substrate

4; luminescence equipment

5; mounting lead

6; inner lead

7; the 1st emitter (350-415nm emitter)

8; the resin section which made the fluorescent substance in this invention contain

9; a conductive wire

10; mould member

[Translation done.]

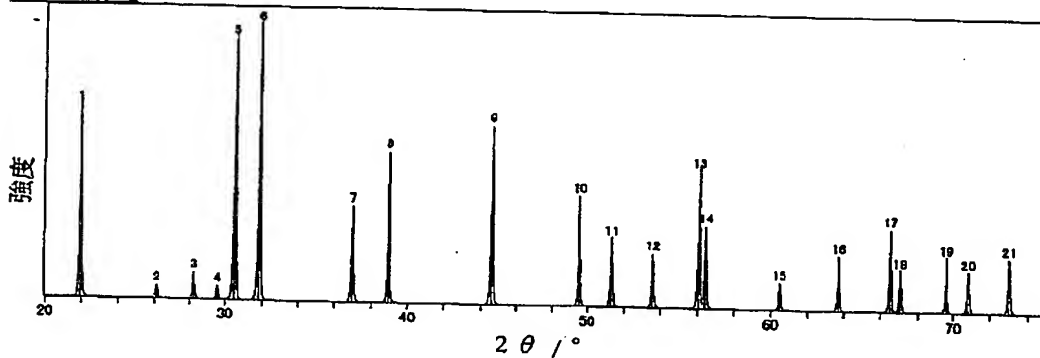
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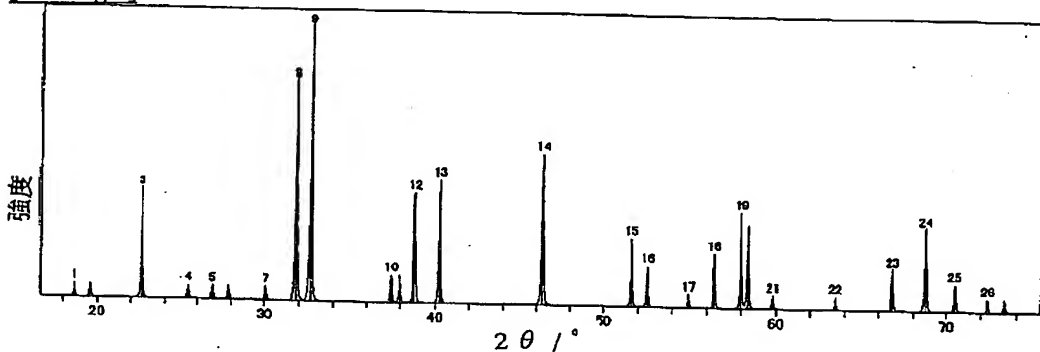
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DRAWINGS

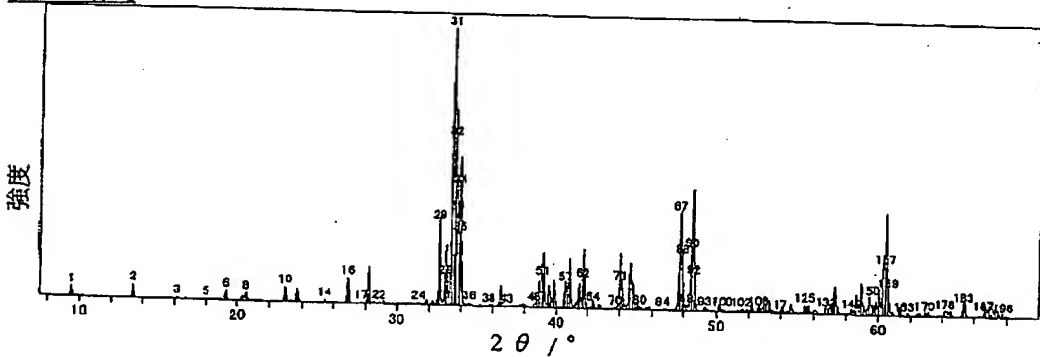
[Drawing 1]



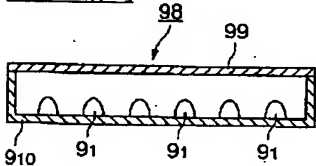
[Drawing 2]



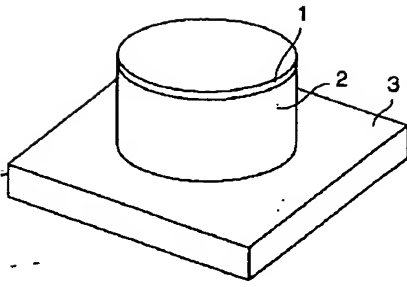
[Drawing 3]



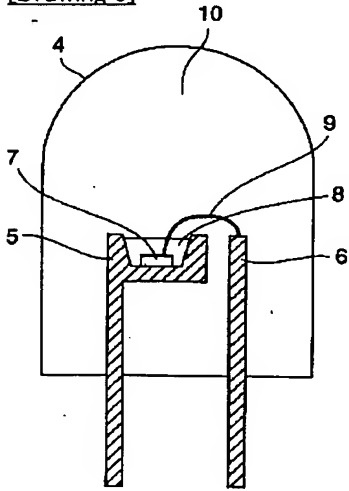
[Drawing 9]



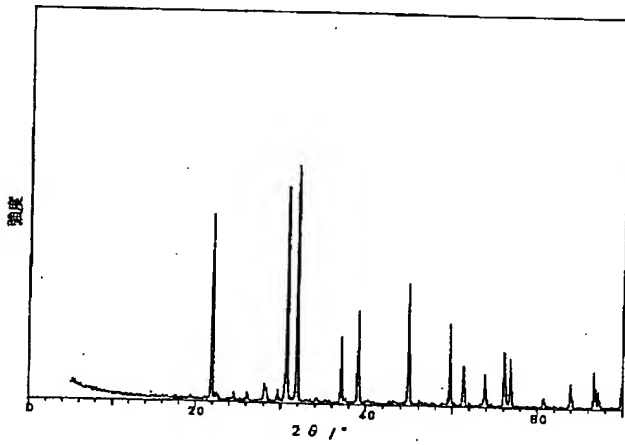
[Drawing 4]



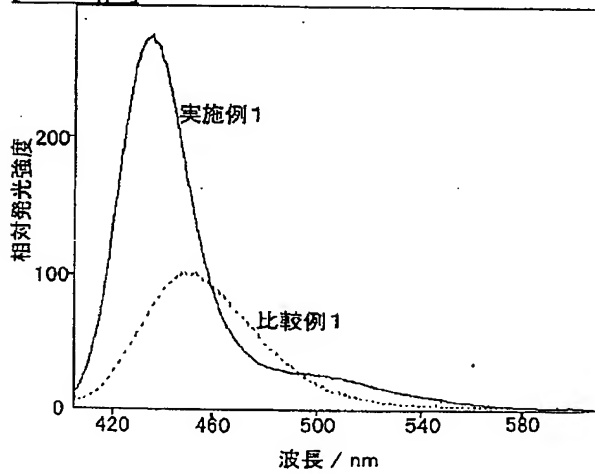
[Drawing 5]



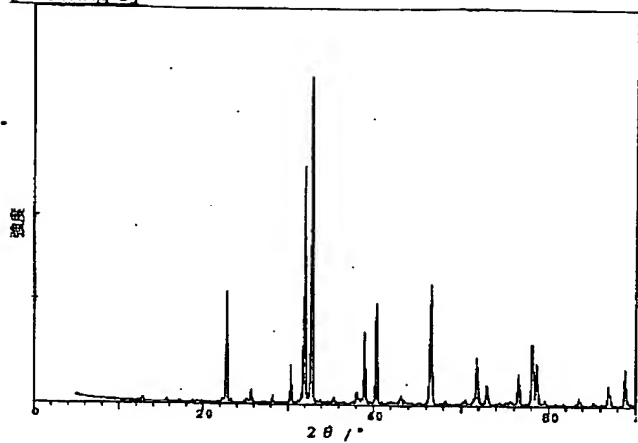
[Drawing 6]



[Drawing 7]



[Drawing 8]



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CORRECTION OR AMENDMENT

[Kind of official gazette] Printing of amendment by the convention of 2 of Article 17 of Patent Law
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H01L 33/00 C
H01L 33/00 N

[Procedure amendment]
[Filing Date] December 9, Heisei 17 (2005. 12.9)
[Procedure amendment 1]
[Document to be Amended] Description
[Item(s) to be Amended] Claim
[Method of Amendment] Modification
[The content of amendment]
[Claim(s)]

[Claim 1] Luminescence equipment characterized by coming to contain the fluorescent substance which has the crystal phase in which said 2nd emitter has the chemical composition of a general formula [1] in the luminescence equipment which has the 1st emitter which generates the light of 350 to 415 nm, and the 2nd emitter which generates the light by the exposure of the light from the 1st emitter concerned.

[Formula 1]

$M1aEubM2cM3dOe \dots [1]$

(However, at least a kind of element chosen from the group which M1 becomes from Ba, Sr, and calcium) the sum total -- more than 90mol% -- the included metallic element is expressed and M2 is chosen from the group which consists of Mg and Zn -- having

** -- even if few -- a kind of element -- the sum total -- more than 90mol% -- the included metallic element -- expressing -- M3 -- Si and G

at least a kind of element chosen from the group which consists of e -- the sum total -- more than 90mol% -- the number with which the included metallic element is expressed and a is satisfied of $2.5 \leq a \leq 3.3$, the number with which b is satisfied of $0.0001 \leq b \leq 1.0$, the number with which c is satisfied of $0.9 \leq c \leq 1.1$, the number with which d is satisfied of $1.8 \leq d \leq 2.2$, and e are numbers with which are satisfied of $7.2 \leq e \leq 8.8$.

[Claim 2] The rate of occupying in M1 of the sum total of Ba, Sr, and calcium, ** of Mg and Zn

Luminescence equipment according to claim 1 with which the rate of occupying in M2 of the total, and the rate of occupying in M3 of the sum total of Si and germanium are characterized by being more than 95mol%, respectively.

[Claim 3] M1 expresses at least one sort chosen from Ba, Sr, and calcium,

Luminescence equipment according to claim 1 or 2 characterized by for M2 expressing at least one sort chosen from Mg and Zn, and expressing at least one sort as which M3 is chosen from Si and germanium.

[Claim 4] ***** given in claim 1 characterized by for M1 expressing at least one sort chosen from Ba and Sr, for M2 expressing Mg, and M3 expressing Si thru/or any 1 term of 3

**.

[Claim 5] Luminescence equipment given in claim 1 characterized by b being $0.001 \leq b \leq 1$ thru/or any 1 term of 4.

[Claim 6] It is the description about the mole ratio of Ba to Sr in M1 being 0.05 or more.

Luminescence equipment given in claim 1 to carry out thru/or any 1 term of 5.

[Claim 7] Luminescence equipment given in claim 1 characterized by the 1st emitter being laser diode or light emitting diode thru/or any 1 term of 6.

[Claim 8] Luminescence equipment according to claim 7 with which the 1st emitter is characterized by being the laser diode or light emitting diode using a GaN system compound semiconductor.

[Claim 9] It is the description about the 1st emitter being surface-emitting type GaN system laser diode.

Luminescence equipment according to claim 8 to carry out.

[Claim 10] Luminescence equipment according to claim 9 characterized by the 2nd emitter being the film-like.

[Claim 11] Efficient luminescence equipment given in claim 1 characterized by making the film surface of the 2nd emitter come to contact the luminescence side of the 1st emitter directly thru/or any 1 term of 10.

[Claim 12] Luminescence equipment given in claim 1 to which the 2nd emitter is characterized by making resin come to distribute the powder of a fluorescent substance thru/or any 1 term of 11.

[Claim 13] The lighting system which has luminescence equipment of a publication in claim 1 thru/or any 1 term of 12.

[Translation done.]